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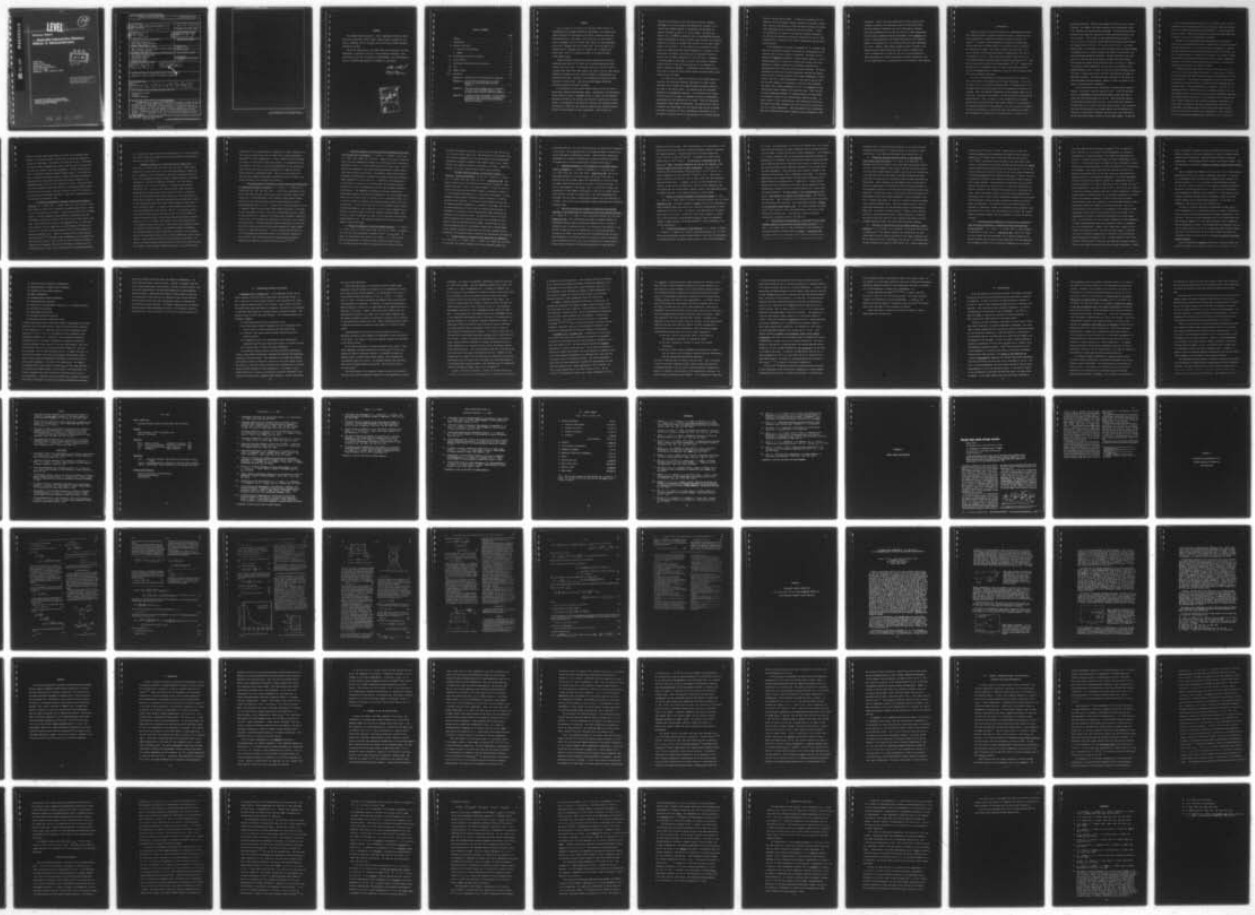

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Summary Report

— Optically Induced Hot Electron Effects in Semiconductors

prepared by
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September 1, 1976 - August 31, 1978

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~~We report here,~~ our progress in using the high electric fields and ultrashort pulses from mode-locked lasers to study the temporal evolution of the saturable optical properties of semiconductors on a picosecond time scale. In these studies, the excite and probe technique is employed to obtain information concerning the ultrafast dynamics of photogenerated electron-hole plasmas. Germanium has been chosen for the initial investigations since its bandgap energy is comparable to but less than the photon energy of the Nd: glass laser.

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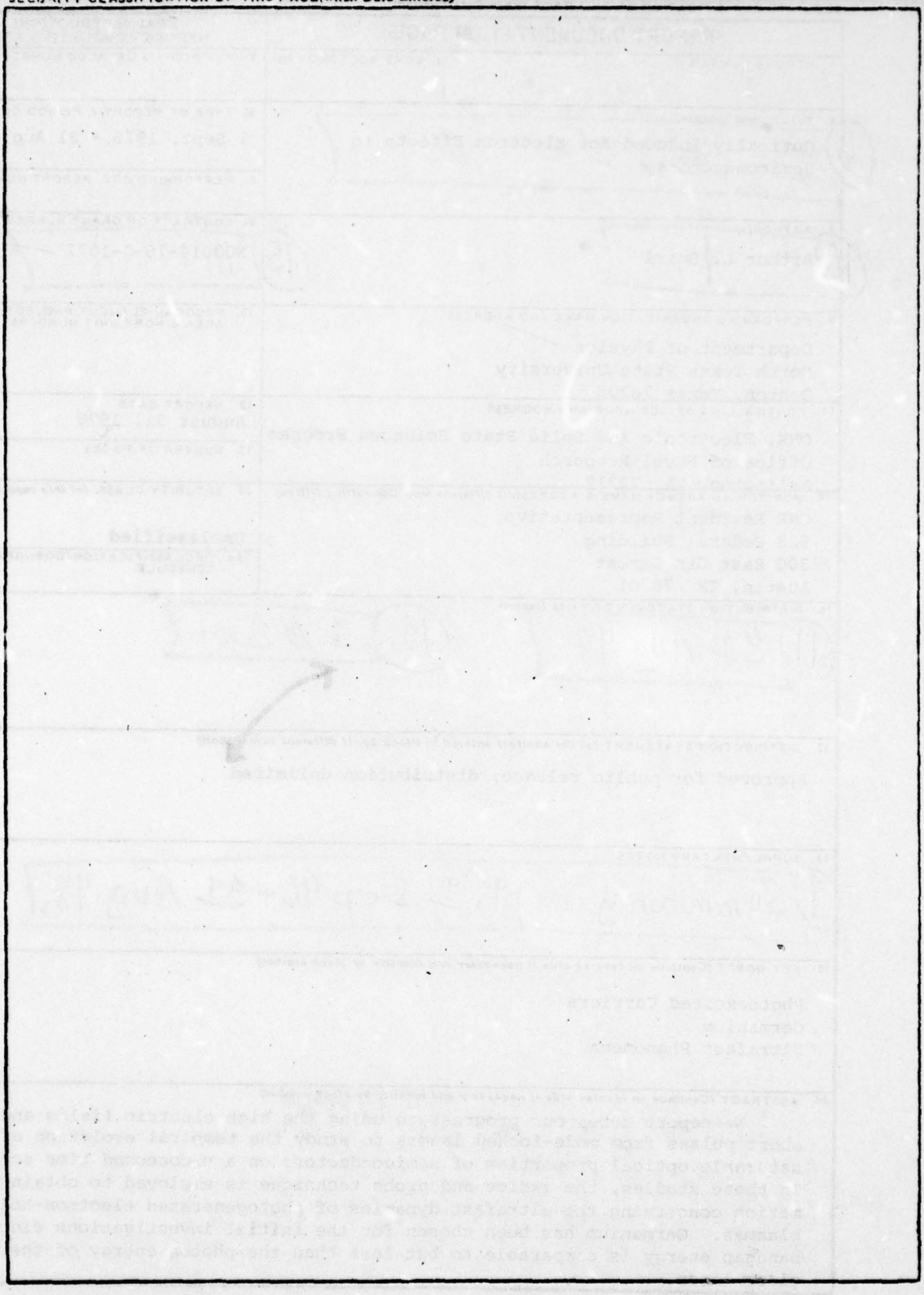

FOREWARD

This Summary Report by Arthur L. Smirl, Department of Physics, North Texas State University, Denton, Texas, covers research progress for the period 1 September 1976 to 31 August 1978, under Office of Navel Research Contract NR 318-048.

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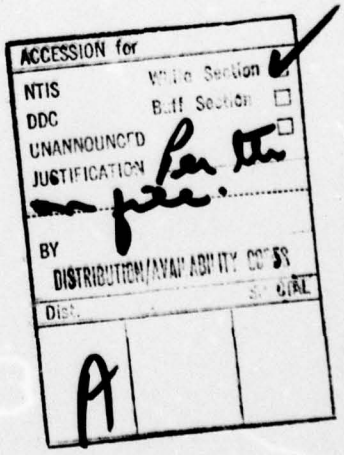
Arthur L. Smirl
Principal Investigator



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SUMMARY

Here, we report on our theoretical and experimental progress in understanding the ultrafast kinetics of high-density, hot electron-hole plasmas generated in semiconductors by intense picosecond pulses from mode-locked lasers. This report covers the entire two year contract period 1 September 1976 to 31 August 1978; however, we have emphasized the period 1 September 1977 to 31 August 1978. The first year of the contract period 1 September 1976 to 31 August 1977 is treated only briefly here, since it is covered in detail in our previous year's annual summary report.

Intense ultrashort optical pulses having durations of a few picoseconds and peak powers of 10^8 watts can be readily generated by mode-locking a Nd: glass laser. The extremely high power and short duration of pulses from these lasers make possible the study of the saturable optical transmission properties and hot electron dynamics of semiconductors on a picosecond time scale. Generally, we have employed the excite and probe technique in these studies. They have provided direct information, concerning ultrafast electronic processes.

At the beginning of the initial contract period, we did not have an operational picosecond spectroscopic system. Consequently, the experimental effort during the period 1 September 1976 to 31 August 1977 was devoted to designing, developing, and ensuring the operational integrity of the basic system. Each component of the system was installed and rigorously tested. The system as a whole was then tested by comparing data obtained using our system with that obtained at other laboratories. Theoretically, we

(together with coworkers at the University of Arizona) completed development of an initial model that describes the evolution of hot electron-hole plasmas produced in germanium by the absorption of intense, ultrashort optical (λ = 1.06 μm) pulses. The model was shown to be in satisfactory agreement with the experimentally-observed enhanced transmission of single, energetic pulses through thin germanium samples as a function of incident pulse energies for two sample temperatures, 105 K and 300 K. It also satisfactorily described the transmission of a weak probe pulse through germanium as a function of time delay after an energetic excitation pulse for different excitation energies and for sample temperatures of 105 K and 300 K. Subsequent to the development of this model, van Driel and coworkers conducted further studies in which the energy band gap of the germanium was tuned by hydrostatic pressures. A modification of our model and computer codes to allow for these effects showed that these studies corroborated the proposed model.

As a result of our numerical evaluation of the proposed model, we found that the theoretical enhanced transmission of single intense, ultrashort pulses through thin germanium samples depended strongly on the width of the optical pulses. We suggested that this pulsewidth dependence be experimentally measured. In collaboration with workers at the University of Arizona, we compared the results of experimental measurements with the predictions of our model. There was substantial disagreement between model and experiment. The question then arose as to whether this disagreement with the proposed model was due to assumptions made in deriving the model, experimental uncertainties in the the physical constants used in the calculations, or to relevant physical

processes omitted from the model. A numerical investigation of the effect of the electron-phonon coupling constants on the model revealed that, given the uncertainties in these and other constants, the pulse-width experiment was not a definitive test for the model. In fact, the uncertainties in certain of the physical constants contained in the calculations prevented our uniquely attributing the rise in probe transmission to a cooling of a hot carrier distribution created by the excitation pulse.

At the beginning of the period 1 September 1977 to 31 August 1978, there was considerable discussion concerning the interpretation of our previous excite and probe studies in germanium. Previously, we had measured the enhanced transmission of single optical pulses at 1.06 μm through germanium as a function of incident pulse energy, and we had measured the temporal evolution of this enhanced transmission on a picosecond time scale using the excite and probe technique. The latter measurements reveal that the probe transmission increases for approximately 100 psec following excitation for a sample temperature of 100 K; however, the rise in probe transmission is less than 40 psec at 300 K. We, and other workers at the University of Arizona, have attributed the slower rise in probe transmission at 100 K to a cooling and a hot electron-hole plasma created by the excitation pulse. Thus, the rise in probe transmission measures the energy relaxation rate and should be a sensitive measure of the electron-phonon coupling constant. In sharp contrast to this interpretation, other workers have stated that they expect the energy relaxation time to be too short to account for the rise in probe transmission. They have suggested that intervalence-band and Coulomb-assisted indirect absorption may be important in such

experiments. Indeed, they have suggested that these processes might introduce a minimum in the absorption versus carrier density curve and have suggested, informally, that this curve might be combined with Auger recombination to account for the rise in probe transmission. We have recently completed detailed measurements of the combined free-carrier, intervalence-band, and indirect absorbance in thin germanium samples at a wavelength of 1.55 μm during excite and probe experiments at 1.06 μm. However, these effects do not introduce a minimum in the absorption versus density curve and, thus, cannot be combined with carrier recombination to account for the rise in probe transmission. These experiments also provide measurement of Auger recombination rates on thin samples that are in agreement with those obtained by other workers.

I. INTRODUCTION

Recently, studies of the optical properties of high-density electron-hole plasmas generated in undoped germanium by intense, ultrashort pulses have provided direct information concerning ultrafast electronic processes.[1-16] Generally, these experimental studies have employed mode-locked pulses from a Nd:glass laser as an excitation source to generate the electron-hole plasma. This source provides pulses that are 5 to 10 psec in duration and that have peak powers of approximately 10^8 watts at a wavelength of 1.06 μm. These pulses when focused on the surface of a thin germanium sample can easily produce a measured irradiance of 10^{-2} J/cm^2 and can create carrier densities of approximately 10^{20} cm^{-3}. Germanium has been chosen as a candidate for study in these investigations primarily because its bandgap energy is comparable to but less than the photon energy of a 1.06 μm Nd:glass laser pulse.

As a rule, the experimental investigations have used a variation of one or the other of two conceptually very simple techniques. In the first of these, a thin (a few microns) germanium wafer is irradiated with intense ultrashort pulses of varying energy, and the transmission of each pulse is recorded. Thus, the nonlinear transmission of the germanium sample can be plotted as a function of pulse energy. The second method is the excite and probe technique. Here, the sample is first irradiated with an intense optical pulse (excitation pulse) that causes a change in the transmission or reflection properties of the germanium. This initial pulse is followed, at various time delays, by a weak probe pulse that measures the change in the transmission or reflectivity of the germanium as it returns to its

equilibrium condition. There are any number of variations on this latter technique. For example, the probe wavelength can be purposefully chosen to be different from the excitation wavelength. We have found this latter variation to be useful for isolating on certain processes during the carrier relaxation. Specifically, we have employed excitation pulses at 1.06 μm (1.17 ev) to generate a dense electron-hole plasma and then probed this plasma both at 1.06 μm (1.17 ev) and 1.55 μm (0.8 ev). The latter photon energy is not sufficient to excite carriers across the direct bandgap in germanium and, as a result, monitors only indirect, free-carrier, and intervalence-band absorption properties. Another variation of this technique is to hold the time delay between excitation and probe constant while varying the energy of the excitation pulse. Of course, in addition to varying the optical parameters, one can vary the environment of the germanium sample as well. To date, we have only varied the sample temperature over a limited range (300 K and 100 K) and subjected the sample to hydrostatic pressure. The excite and probe technique is also useful for stimulated-Raman-scattering studies; however, to date, the author is not aware of any such studies in germanium.

As we have mentioned, these techniques are embarrassingly simple in concept. In practice, quite the opposite is true. First, the Nd:glass laser is a temperamental device providing optical pulses that vary in energy and width from one pulse to the next. These pulses are usually of questionable spatial quality as well. Add to these laser properties the facts that one is trying to maintain spatial overlap of the excitation pulse and the probe pulse on the sample surface (each focused spot is a fraction of a millimeter in diameter) and that one is often operating at optical intensities within a factor of two of sample damage. In addition,

if a probe wavelength different from 1.06 μm is desired, then it must be generated by some nonlinear process such as frequency doubling, frequency tripling, or Raman scattering--we have employed all of these methods and none are trivial. Thus, data acquisition on these time scales employing a Nd:glass laser can be an exasperating procedure.

Because of the huge carrier densities generated and the complex nature of the germanium band structure, interpretation of these experiments has been tedious as well. By its very nature, the problem is a complex many-body problem involving multiple processes--in fact, as we shall see, up to sixteen processes or effects may be involved. As a result of these experimental and theoretical problems (and others we haven't mentioned), progress in this field has been painstaking and tedious. None-the-less, progress has been slowly but steadily achieved.

Although there have been reviews of work in this area, they are all over a year old and now grossly out of date. In view of this, I believe it would be useful for reader and author alike to briefly review the relevant literature. I have chosen to accomplish this goal by listing each reference by title and author(s), followed by a short paragraph relating the points that I think are important to this report. This list is not comprehensive but is restricted to work in germanium related to our own. The comments reflect my own point of view and are by definition subjective. This paper-by-paper review (Sec. II) is followed by a few "state-of-the-field" remarks concerning areas of general agreement, controversy, and future study. Section III then gives a chronological record of our achievements and progress during this two year period, and Section IV details our plans for the coming year's experiments. Next, we provide a list of professional publications and activities (Sec. V) during the

past two years. In Section VI, we provide the reader with an updated vita, and in Sec. VII we provide a budget summary for the previous two year period. Finally, in Appendices A, B, C, and D, we provide reprints and preprints of our work during the period 1 September 1977 to 1 September 1978. Reprints for the first contract year, 1 September 1976 to 1 September 1977, are contained in our last year-end report.

II. SURVEY OF THE FIELD

Review of the Literature

In this section we provide a paper-by-paper account of the progress in our understanding of the temporal evolution of optically-generated electron-hole plasmas on a picosecond time scale. I believe such a chronological listing is useful in providing some perspective as to why certain investigations were undertaken. It also allows me, in light of our more recent studies, to make a few arbitrary comments concerning some of these works.

1. "Nonlinear Absorption and Ultrashort Carrier Relaxation Times in Germanium Under Irradiation by Picosecond Pulses," C. J. Kennedy, J. C. Matter, A. L. Smirl, H. Weichel, F. A. Hopf, S. V. Pappu, and M. O. Scully, Phys. Rev. Lett. 32, 419 (1974). This paper reports the results of measurements performed by Smirl and Matter in which they observed a saturation of the absorption of 1.06-μm picosecond pulses in thin germanium wafers at high optical intensities. They performed two experiments. In the first, they irradiated the 8-μm-thick sample with varying intensity picosecond pulses at 1.06 μm, and they measured the transmission of each pulse. A plot of germanium transmission versus incident optical pulse energy showed that the germanium transmission was bleached or enhanced by a factor approximately 20 over its linear value at low intensities. Second, these authors employed the excite and probe technique in an attempt to measure the decay of this enhanced transmission in the following manner. They irradiated the sample with an excitation pulse intense enough to bleach the sample transmission by a factor of ~20. This excitation pulse was then followed, at various

delays, by a probe pulse that measures the decay of the enhanced transmission. The authors observed a narrow spike in the probe transmission located near zero delay. The width of the spike was <5 psec. No further structure in probe transmission was seen at this time due to the nature of the excitation intensity and laser performance. Further structure would later be reported by Shank and Auston[3] and Smirl et al.,[5] as we shall discuss. In absence of further structure, however, the authors erroneously interpreted this narrow probe spike near zero delay as evidence of an intraband relaxation time for hot electrons of <5 psec. This spike would later be interpreted by Shank and Auston[3] as a correlation effect between probe and excitation pulses, and more recently, by Ferry[14] as being due to state filling and band-gap narrowing. These later interpretations will be discussed as we encounter them.

2. "Picosecond Ellipsometry of Transient Electron-Hole Plasmas in Germanium," D. H. Auston and C. V. Shank, Phys. Rev. Lett. 32, 1120 (1974). Here, Auston and Shank first illuminated a germanium sample with an intense 1.06 μm picosecond pulse to produce a large carrier density near the sample surface. They then employed a weak reflected 1.06-μm-probe pulse and ellipsometric techniques to monitor the decay of this carrier density with time. They attributed the decay of this surface density to a diffusion of the carriers into the sample bulk, and they deduced a diffusivity of 230 cm^2 sec^{-1} (a value 3.5 times larger than the low density ambipolar diffusion constant in Ge). However, as later studies by Auston et al.[4] and Smirl et al.[13,16] would show, Auger recombination can be important at these densities as well. Thus, we believe that the diffusion constant deduced on the assumption that the carrier density is depleted only by diffusion may be in error. One of the goals of our ongoing measurements

is to separate the Auger and diffusion contributions to the carrier density decay and to deduce exact numbers for carrier recombination and diffusion constants at these densities.

3. "Parametric Coupling in an Optically Excited Plasma in Ge," C. V. Shank and D. H. Auston, Phys. Rev. Lett. 34, 479 (1975). These authors repeated the 1.06 μm excite and 1.06 μm probe measurements of Kennedy et al.[1] In addition to the narrow spike in the probe transmission near zero delay, the measurements revealed a slower, broader structure in the probe transmission. The probe transmission exhibited a slow rise lasting approximately 20 to 30 psec followed by a gradual decrease lasting hundreds of psec. In view of this additional data, Shank and Auston reinterpreted the narrow probe spike as being the result of scattering of the strong excitation beam into the direction of the probe beam by a phase grating formed by a spatially modulated index of refraction created by the interference between the strong and weak beams--in other words, it was a correlation effect. In addition, they attributed the slower rise in probe transmission to band filling. That is, to a filling of conduction (valence) band states by electrons (holes) to the point where the separation between electron and hole Fermi energies approaches the photon energy 1.17 ev. As a result, the buildup of this effect should follow the integrated optical pulse energy. Notice that this interpretation does not invoke hot electron effects. The electrons (holes) are assumed to have already relaxed to lattice temperature. According to this interpretation, the correlation spike and rise in probe transmission contain little physics. They are merely artifacts of the measurement techniques: one being a correlation between excitation pulse and probe pulse, and the other, the integral of the intensity correlation function. As we have recently

pointed out,[16] this interpretation is adequate for a sample temperature of 300 K, but it cannot account for the slower rise in probe transmission observed[5,16] at lower temperatures. In fact, Ferry[14] has even questioned the origin of the so called "correlation spike". As we progress through the literature, we shall see that the interpretation of the "correlation spike" and the slow rise in probe transmission as a function of delay become sources of considerable debate. This is not surprising since these are very complex interactions involving huge electric fields, large carrier densities, and a complex band structure.

4. "Picosecond Optical Measurements of Band-to-Band Auger Recombination of High-Density Plasmas in Germanium," D. H. Auston, C. V. Shank, and P. LeFur, Phys. Rev. Lett. 35, 1022 (1975). In this novel application of the excite and probe technique, Auston et al. first illuminated a slab of germanium with a 1.06-μm-excitation pulse creating a large carrier density. The decay of this carrier density was then probed employing a second Raman-generated probe at 1.55 μm. A quanta of the excitation pulse (1.17 ev) is energetic enough to excite direct band-to-band transitions. The energy of the probe quanta (0.8 ev) is less than the energy-band gap and is taken by these authors to be a measure of the free-carrier absorption coefficient. A plot of the 1.55-μm-probe pulse transmission versus delay, thus, displays the carrier decay. The probe absorbance decays significantly in the first 100 psec following excitation indicating the importance of Auger recombination on a picosecond time scale. It must be emphasized here that the 1.55 μm absorbance was attributed entirely to free-carrier transitions. We now believe that intervalence-band and Coulomb-assisted indirect absorption are important at these wavelengths as well.[12,13,16]

5. "Ultrafast Relaxation of Optically Excited Nonequilibrium Electron-Hole Distributions in Germanium," A. L. Smirl, J. C. Matter, A. Elci, and M. O. Scully, Opt. Commun. 16, 118 (1976). Unaware of the work of Shank and Auston,[3] we had independently extended our 1.06-μm-excite and 1.06-μm-probe experiments of Ref. 1 to include probe structure at longer delays. Here, we reported the dependence of the excite-probe measurements on sample temperature and excitation energy levels. Specifically, single pulse nonlinear transmission was measured as a function of incident optical pulse energy at sample temperatures of 105 K and 297 K. In addition, the transmission of a probe pulse as a function of time delay after an excitation pulse was measured for the same two temperatures and for three different excitation pulse energies. The temperature dependence of the probe transmission measurements contained surprising new information: the rise in probe transmission at 105 K was too slow (100 psec) to be an integration effect (i.e., it could not simply be following the integrated optical energy of the excitation pulse). It was in this work that we first proposed a model, based on the above measurements, that attributed the rise in probe transmission to a cooling of electrons (holes) within the conduction (valence) bands. Thus, the rise in probe transmission was taken to be an indication of the carrier energy relaxation time. The theoretical model was still very crude and numerical curves were rough.

6. "Saturable Transmission in Mercury Cadmium Telluride," J. C. Matter, A. L. Smirl, and M. O. Scully, Appl. Phys. Lett. 28, 507 (1976). We applied the excite and probe measurement techniques of Ref. 5 to $Hg_{1-x}Cd_xTe$. We chose this semiconductor for study for two reasons: (1) it was a direct band gap semiconductor, providing a simpler theoretical model than Ge. (2) its band gap could be adjusted by varying the mole fraction of CdTe.

For these measurements we chose an x value of 0.61 so that the band gap (0.75 ev) was close to that of Ge. The results were very different from the Ge results--probably due to the polar nature of the electron-phonon interaction in HgCdTe as opposed to the nonpolar nature in Ge. Experimentally, the material was next to impossible to work with. Because the material was so soft, damage occurred easily during polishing.

7. "Ultrafast Transient Response of Solid-State Plasmas. I. Germanium: Theory and Experiment," A. Elci, M. O. Scully, A. L. Smirl, and J. C. Matter, Phys, Rev. B 16, 191 (1977). Supported by ONR. This paper presents the only first-principles theoretical treatment of these problems available to date. This model attempts to account for the nonlinear single pulse and excite-probe response of germanium in terms of: (1) direct band-to-band absorption, (2) free-carrier absorption, (3) long wavevector phonon-assisted intervalley carrier scattering, (4) phonon-assisted carrer relaxation, (5) carrier-carrier Coulomb collisions and (6) plasmon-assisted recombination. According to this model, the rise in the probe transmission with time after an intense excitation pulse is attributed to a cooling of the hot electron-hole plasma created by the absorption of the excitation pulse. The theoretical fits to the single pulse transmission date and probe pulse date can be regarded as satisfactory, given the complexity of the problem. None-the-less, we noted from the beginning that our calculations included only a limited number of the possible electronic interactions and that they contained serious assumptions that warranted further theoretical and experimental investigation.

8. "Pressure Tuning of Picosecond Pulse Transmission in Germanium," H. M. van Driel, J. S. Bessey, and R. C. Hanson, Opt. Commun, 22, 346 (1977). In this work, the authors irradiated thin ($\sim 5\mu m$) germanium samples with

picosecond pulses at 1.06 μm of varying intensity and measured the transmission of each pulse. The energy band gap of the germanium sample was tuned by subjecting the sample to hydrostatic pressure. These nonlinear transmission measurements as a function of hydrostatic pressure followed and confirmed the predictions of the hot electron model proposed by Elci et al.[7]

9. "Pulsewidth Dependence of the Transmission of Ultrashort Optical Pulses in Germanium," J. S. Bessey, B. Bosacchi, H. M. van Driel, and A. L. Smirl, Phys, Rev. B 17, 2782 (1978). Supported by ONR. One of the interesting features of our initial model (Elci et al.[7]) was that it predicted that the nonlinear single pulse transmission of the thin germanium sample should strongly depend on the width of the optical pulses. In fact, we had suggested in Elci et al.[7] that, as a test for our model, the transmission of these samples be measured as a function of incident optical pulse energies for pulses of various width. Here such measurements were performed; they were found to substantially disagree with the predictions of our model. This was the first disagreement between experiment and a heretofore successful model.

10. "The Role of Phonons and Plasmons in Describing the Pulsewidth Dependence of the Transmission of Ultrashort Optical Pulses Through Germanium," W. P. Latham, Jr., A. L. Smirl, A. Elci, Solid-State Electron. 21, 149 (1978). Supported by ONR. This investigaiton was begun to determine whether the disagreement between theory and experiment reported in Ref. 9 was due to assumptions made in deriving the model, the experimental uncertainties in the physical constants used in the calculations, or the physical processes included in the model. The result was that we demonstrated that the pulsewidth dependence of the plasmon effects is opposite to that of the phonon-assisted relaxation and that, due to the uncertainties in electron-optical phonon coupling constants, the pulsewidth experiments did not provide a

definitive test of the model. This investigation of the uncertainties in the known vaules of the electron-phonon coupling constants produced an important and disquieting result: accepted values of this constant ranged from 6.4×10^{-4} erg-cm^{-1} to 18.5×10^{-4} erg-cm^{-1}. An average of the eight values reported here is 1×10^{-3} erg-cm^{-1}. If this value is substituted into the Elci et al.[7] model, the energy relaxation rate for the hot carriers is too rapid to account for the rise in probe transmission. Elci et al.[7] had originally used electron-phonon coupling constants of 6×10^{-4} erg-cm^{-1} at a lattice temperature of 297 K and 2×10^{-4} erg-cm^{-1} at 100 K. These values result in carrier rates that are 3 and 25 times slower than that obtained by using the average value. In summary, the rise in probe transmission with time may, or may not, be accounted for by carrier cooling depending on the value chosen for the electron-phonon coupling constant; more experimental data is required (see Ref. 15 for further discussion of these points).

11. "Physics of Ultrafast Phenomena in Solid-State Plasmas," A. Elci, A. L. Smirl, C. Y. Leung, and M. O. Scully, Solid-State Electron. 21, 151 (1978). Supported by ONR. This paper presents an invited review of our experiments in which picosecond pulses were used and our theoretical understanding of these experiments as of July, 1977. It contains a good description of our experimental work to this point. In addition to our review, however, we removed one of the restrictions of our earlier Elci et al.[7] model by considering through a simple analysis, the possible effects of carrier diffusion on our optical measurements.

12. "Picosecond Spectoscopy of Semiconductors," D. H. Auston, S. McAfee, C. V. Shank, E. P. Ippen, and O. Teshke, Solid-State Electron. 21, 147 (1978). These authors presented excitation-probe studies in GaAs that were interpreted as demonstrating that the energy relaxation rate in this polar semiconductor

was ~2 psec. More importantly, at least for our purposes here, these authors stated that they expected the energy relaxation times in nonpolar germanium to be too short to account for the rise in probe transmission. This suggestion is, of course, consistent with our more detailed calculations presented in Latham et al.[10] Furthermore, they suggested that enhanced intervalence-band and Coulomb-assisted indirect absorption effects might be important at the high optically-created carrier densities encountered in our recent excite and probe experiments. These processes might introduce a minimum in the absorption versus density curve in germanium, they claimed. In a private communication and in remarks before the International Conference on Hot Electron Effects in Semiconductors, S. McAfee and D. H. Auston further explained how such an absorption curve could then be combined with Auger recombination to explain the rise in probe transmission in recent excite and probe experiments. In sharp contrast to the interpretation of Elci et al.,[7] this model did not require hot electron effects. The principal processes were: Auger recombination, Coulomb-assisted indirect absorption, and intervalence-band absorption. This second model required a minimum in the absorption versus density curve. Our studies presented in Latham et al.[10] had convinced us that the interpretation of the rise in probe transmission was again open. This model was indeed an attractive alternative--we decided to test it (see Ref. 16).

13. "Picosecond Optical Absorption at 1.06 μm and 1.55 μm in Thin Germanium Samples at High Optically-Created Carrier Densities." A. L. Smirl, J. R. Lindle, and S. C. Moss, Proceedings of the Conference on Picosecond Phenomena, Springer-Verlag (in press). Supported by ONR. Preliminary results and conclusions of our attempts to measure the contributions of (1) Auger recombination, (2) Coulomb-assisted indirect

absorption and (3) intervalence-band absorption to our excite and probe experiments were presented. We shall postpone any discussion of these results until a more complete presentation can be given in Ref. 16.

14. "Energy-Gap Narrowing and State Filling in Semiconductors Under Intense Laser Irradiation," D. K. Ferry, to be published. To appreciate this work one needs to recall two facts from previous studies: (1) that a major assumption of Elci et al.[7] was that the carrier-carrier collision rate was high enough to justify taking the carrier distributions to be Fermi-Dirac and (2) that Shank and Auston[3] attributed the narrow spike near zero probe delay to correlation effects between excitation and probe pulses. Ferry reexamines the approximation that the distribution function is Fermi-Dirac by calculation the time and energy dependence of the distribution function at the high carrier photogeneration rates encountered here. He concludes that on a time scale of tens of picoseconds the distribution function does indeed approximate a Fermi distribution; however, on shorter time scales, it contains a δ-function-like spike located at the optically coupled states. Thus, for purposes of calculating the probe pulse transmission, one may reasonably assume the distribution is Fermi-like. In contrast to Shank and Auston,[3] however, Ferry concludes that the so called correlation spike in probe transmission is caused by the state filling that results in this δ-function-like spike in the distribution function on short time scales and by band-gap narrowing.

15. "Influence of Hot Phonons on Carrier Energy Relaxation in Semiconductors," H. M. van Driel, to be published. As we recall from the work of Latham et al.,[10] the physical constants for germanium are not well enough known to allow a precise calculation of the energy relaxation rate. For the theoretical fits to experiment by Elci et al.,[7] the electron-phonon coupling constants were chosen as 6×10^{-4} erg-cm^{-1} for a lattice temperature

of 297 K and 2×10^{-4} erg-cm^{-1} at 100K. These values are within the accepted theoretically and experimentally determined values listed by Latham et al.;[10] however they are much lower than the mean value of 1×10^{-3} erg-cm^{-1} as obtained from an average of the eight values listed. In fact, a repetition of the original calculations substituting the average electron-phonon coupling constant shows (see Latham et al.,[10] Fig. 8) that carrier cooling is too rapid to account for the rise in probe transmission, in complete agreement with the claims by Auston et al.[12] However, van Driel calculates the influence of hot phonons on the carrier energy relaxation rate in these problems. These calculations suggest that the long equilibration time for the hot carriers is due to a relaxation bottleneck produced by a buildup of the optical phonon population on a picosecond time scale. These calculations, taking into account optical phonon heating and employing only a single average temperature-independent electron-phonon coupling constant, removes the major discrepancies between our original theory and experiment. The agreement between the modified theory and experiment is remarkable; however, this agreement must be regarded as somewhat fortuitous in view of the simplifications of the model, the limited number of processes included, and the uncertainty in many of the physical constants.

16. "Picosecond Optical Measurement of Free-Carrier, Intervalence-band, and Indirect Absorption in Germanium at High Optically-Coupled Carrier Densities." A. L. Smirl, J. R. Lindle, and S. C. Moss, accepted for publication by Phys. Rev. B. Supported by ONR. Recall that amoung our early studies were the measurement of the enhanced transmisison of single ultrashort optical pulses through germanium[1,5] and the measurement of the temporal evolution of this enhanced transmission on a picosecond

time scale using the excite and probe technique.[3,5] As the reader will recall (see Ref. 5), in the first of these experiments, the nonlinear transmission of a single picosecond 1.06 μm pulse was investigated as a function of incident optical pulse energy for sample temperatures of 100 K and 297 K. In the second study, the sample was first irradiated by an excitation pulse of sufficient energy to cause the transmission of the germanium to be enhanced. This initial pulse was then followed at various time delays by a weak probe pulse, of the same wavelength, that measured the temporal evolution of the germanium transmission. The latter measurements reveal that the probe transmission increases for approximately 100 psec following excitation for a sample temperature of 100 K; however, the rise in probe transmission is less than 40 psec at 297 K. We have seen that there is a great deal of debate concerning this rise in probe transmission. Originally, Shank and Auston[3] attributed this rise in the probe transmission to a saturation of the absorption caused by a filling of the optically-coupled conduction-band states and a depletion of the valence-band states by direct band-to-band transitions induced by the excitation pulse. Thus, the optically-created carrier density, and consequently the increase in probe transmission, should follow the integrated optical pulse energy. This interpretation was based on observations performed only at room temperature. As we show here, the rise in probe transmission at room temperature is indeed indistinguishable from other integration effects, in agreement with this interpretation. However, such a model cannot account for the slower rise observed at 100 K. We recall that Elci *et al.*,[7] attributed this rise in the probe transmission to a cooling of a hot electron-hole plasma created by the excitation pulse. In sharp contrast to this interpretation, Auston *et al.*,[12] stated that they expected the energy relaxation time to be too short to account for the rise in probe transmission. Indeed,

Auston and McAfee[12] suggested a plausible alternative explanation for the temporal evolution of the probe transmission in terms of enhanced Coulomb-assisted indirect absorption, intervalence-band absorption, and Auger recombination. This explanation did not require hot electron effects. This last explanation did require a minimum in absorption versus density relationship.

Thus, we summarize and emphasize that there are at least three possible explanations for the rise in probe transmission with delay: (1) the rise is an intergration effect following the integrated optical energy of our excitation pulse--i.e., it is an artifact of the measurement technique, (2) the rise is due to a cooling of a hot carrier distribution created by absorption of the excitation pulse, or (3) the rise can be attributed to Auger recombination combined with an absorption vs. carrier density relationship containing a minimum, where the minimum is caused by Coulomb-assited indirect and intervalence-band absorption. In this work, we attempted to test the first and third possiblilites.

We began by carefully repeating the measurements by Smirl et al.[5] of the transmission of a 1.06-μm-probe pulse as a function of time delay after an intense 1.06-μm-excitation pulse for sample temperatures of 100 K and 295 K. The rises in probe transmission for the two temperatures were then carefully compared to a calculated intergration curve, assuming an optical pulsewidth of 10 psec. We concluded from this comparison that the rise in probe transmission at 295 K is indistinguishable from the integration curve, but that the rise at 100 K is much slower than either the integration curve or the rise at 295 K and is not an artifact of the measurement technique.

Next, we measured the combined free-carrier, intervalence-band, and indirect absorbance in thin germanium samples at a wavelength of 1.55 μm

during excite and probe experiments at a wavelenght of 1.06 μm. Our interests in these measurements were twofold. First, we wanted to ascertain whether or not free-carrier, intervalence-band, and indirect absorption effects are important in excite-probe experiments at 1.06 μm. Second, if these effects are important, can they, together with Auger effects, account for the rise in probe transmisison.

In the first of these, the transmission of a thin germanium sample at 1.55 μm and 1.06 μm was measured as a function of optically-created carrier densities. Over the range of densities encountered in these experiments, the absorption versus density relationshop at 1.17 ev did not exhibit a minimum. Thus, a temporal decay of carrier density alone cannot be combined with this absorption versus density relationship to account for the rise in probe transmission at 1.06 μm exactly as suggested by Auston and McAfee. In addition, these measurements indicated that the combined free-carrier, intervalence-band, and indirect absorbance changes are opposite in sign and smaller in magnitude than the changes caused by saturation of the direct absorption. As a result, we believe that the decrease in absorbance at 1.06 μm with increasing carrier number is dominated by a saturation of the direct absorption coefficient; however, the rate of this decrease in absorbance is slowed by the contributions of these "other" processes that are opposite in sign.

In addition, the absorbance of a 1.55-μm-probe pulse was measured as a function of time delay after an intense 1.06-μm-excitation pulse. The 1.55-μm-probe absorbance decayed by approximately 0.8 in the first 100 psec following excitation, corresponding to a transmission increase of a factor of approximately 2. This represents a significant decay in the combined free-carrier, intervalence-band, and indirect absorbance during this period. Contrary, however, to measurements of the probe rise at 1.06 μm,

the decay of probe absorbance at 1.55 μm exhibited no strong dependence on sample temperature.

As a result of the present measurements, we feel that free-carrier, intervalence-band, and Coulomb-assisted transitions combined with Auger recombination are not the mechanisms dominating the rise of 1.06-μm-probe transmission at 100 K. The contributions of these processes are significant, however, and they must be accounted for by any successful model. Unfortunately, these measurements yield no direct information concerning carrier distribution temperatures or energy relaxation rates, and the question of attributing the rise in 1.06-μm-probe transmission to a cooling of a hot carrier plasma created by the excitation pulse remains unresolved.

Remarks

In this section, I would like to make some concluding remarks concerning our review of the field. The remarks mainly concern the remaining problems that I hope we can solve in the immediate future. Again, I remind the reader that the opinions reflect my own bias.

The most obvious observation to be made after our survey of the literature concerns the large number of processes or effects that have been shown to be important by experimentalists or invoked by theorists in these problems. There are at least 16:

(1) Direct band-to-band transitions

(2) Long-wavevector phonon-assisted carrier scattering from central to side valleys

(3) Phonon-assisted indirect band-to-band transitions

(4) Carrier-carrier collisions

(5) Free-carrier absorption

(6) Plasmon-assisted nonradiative recombination

(7) Phonon-assisted intraband carrier relaxation

(8) Diffusion at high carrier densities

(9) Auger recombination

(10) Coulomb-assisted indirect absorption

(11) Intervalence-band absorption

(12) Artifacts of the measurement technique, e.g., correlation effects

(13) Hot phonon distributions

(14) Energy-band gap narrowing

(15) Non-Fermi distribution function

(16) Enhanced radiative recombination rates

I believe that every process listed above occurs to some degree or the other during our excite and probe studies. Some occur during optical excitation at any intensity (1-5, 7), some occur only at high photogenerated carrier densities (6, 8-11, 13, 14, 16), and others are only a byproduct of the measurement technique (12). Obviously, a first principles calculation incorporating all of the above is prohibitive. Moreover, the physical coupling constants, in most cases, are not well enough known to allow such a calculation--even if we were foolish enough to attempt one. How then do we proceed? Or is it worth proceeding at all? It seems to me that, if we are ever to unravel the various roles played by the processes listed above, we must refine our experimental techniques so that we may focus on the processes separately. This effort has already begun. For example, the measurements by Auston et al.[4] and Smirl et al.[16] have been successful in separating the combined effect of free-carrier, intervalence-band, and indirect absorption and Auger recombination from other effects such as direct absorption. Similarly, Auston and Shank[2] have attempted to determine the effects of diffusion at high carrier densities--although these measure-

ments were probably affected by Auger and Radiative recombination. The yield from such studies could be exciting. Suppose, for example, that the rise in probe transmission could be shown to be due to carrier cooling. The probe rise could provide a very sensitive measure of the electron-phonon coupling constant. Another exciting aspect of these studies is that many of the processes listed above are not observed at lower carrier densities, and those that are may have their interactions altered by screening and other effects at these densities. For example, expressions for low density radiative band-to-band recombination may have to be drastically altered at these densities. Our future plans in this area are detailed in Section IV.

III. CHRONOLOGICAL RECORD OF ACTIVITIES

1 September 1976 - 31 August 1977. At the beginning of this initial contract period, we did not have an operational spectroscopic system. In fact, a portion of the local funds required to fully capitalize this system were pledged as matching funds to this contract. Thus, during this period, the experimental effort was devoted to designing, constructing, and assembling a system that would enable us to directly monitor ultrafast processes. This system utilizes an excite and probe technique. This technique requires the following equipment:

(1) A mode-locked laser producing ultrashort pulses.

(2) An electro-optic shutter to separate one of the ultrashort pulses from the train of pulses produced by the mode-locked laser.

(3) A detector - fast scope system to monitor the pulse train and shutter operation.

(4) An apparatus to split the "switched-out" pulse and precisely temporally delay one part (probe) with respect to the other (excite).

(5) An optical dewar to control the sample temperature.

(6) A detection system to monitor the excite and probe transmissions.

In short, this period was devoted to developing and ensuring the operational integrity of the basic system. Each component of the system was installed and rigorously tested (as described in detail in last year's annual report). The system as a whole was tested by comparing data obtained using our present system with data obtained at other laboratories. A by-product of this building effort was a letter in the Review of Scientific Instruments[17] describing our unique optical integrating peak detectors. Further experimental

work will be described below.

During this same period, our theoretical activity centered about providing a detailed theoretical description of the transmission of ultrashort optical pulses through germanium. At the beginning of the year, an initial model had been developed by A. Elci, A. L. Smirl, J. C. Matter, and M. O. Scully, and Smirl and Matter had completed their initial excite-probe experiments in germanium. In addition, an attempt had been made to generate numerical solutions to the complicated set of integro-differential equations in the theory. However, there were several problems with the approximations contained in the model and in the original computer codes. The complete program was lengthy and expensive to execute, and the solution to the differential equations was crude. Several modifications were needed to improve the accuracy and execution time (details in FY 1976-77 annual report).

Having developed an efficient and accurate computer routine, we generated theoretical curves comparing the experiments of Smirl and Matter[5] to our theory. The results of theory and numerical analysis were presented in the Physical Review.[7]

By the time we completed the theoretical paper by Elci et al.,[7] additional experiments had been performed by van Driel et al.[8] in which the bandgap was tuned by hydrostatic pressure. We modified our model and computer codes to include these pressure effects, and we generated the corresponding theoretical curves. The agreement between these new experiments and the proposed model was excellent. The curves are shown in van Driel et al.[8]

One of the results of our numerical studies was that the theoretical nonlinear single pulse transmission exhibited a strong dependence on optical

pulsewidth. As a result, we suggested[7] experimental studies of this type as a test for our model. Since the apparatus at NTSU was not fully operational, A. L. Smirl collaborated with J. S. Bessey, B. Bossachi, and H. M. van Driel at the University of Arizona to conduct single pulse transmission studies in which the incident optical pulsewidth was varied. Theoretical curves for this study were generated at North Texas. The results of these studies were reported in Physical Review,[9] and were discussed during our review of the literature in Sec. II. The pulsewidth measurements indicated the first disagreement between theory and experiment. We then numerically investigated whether this disagreement was due to the simplifying approximations made in the model, the uncertainties in physical constants, or the physical processes included in the model. By numerically solving the equations while varying the physical constants over their accepted range, we generated a large library of figures in which theory and experiment were compared. As a result of this study, we realized that the pulsewidth experiments did not provide a definitive test of our model. Indeed, we realized that the uncertainty in the electron-phonon coupling constant prevented our uniquely attributing the rise in probe transmission to a cooling of the hot carrier distribution created by the excitation pulse. One could reasonably argue that the energy relaxation rate was far too rapid to account for this rise. However, it also became clear to us that if the rise were due to a cooling of a hot carrier distribution then the slope of this rise should be a sensitive measure of the electron-phonon coupling constant. The culmination of this theoretical study was the paper by Latham, Smirl, Elci, and Bessey.[10]

Finally, an invited review paper, in which we included diffusion in our model for the first time, was presented at the International Conference

on Hot Electrons in Semiconductors. These studies showed that, depending on the initial carrier number and spatial distribution immediately after the passage of the excitation pulse, carrier diffusion could lead to a slow (hundreds of psec) decrease or increase in probe transmission. We had originally thought that diffusion might account for the gradual observed decrease in probe transmission after the initial rise.

In summary, although experimentally this was a building year, we provided the theoretical and numerical support needed to compare theory with experiment--not only at NTSU but at the University of Arizona and the University of Toronto as well. We also collaborated on experimental work at Arizona, while assembling our own equipment. We were active professionally. As a result of our work in this area, we published five papers,[7,9,10,11,18] gave an invited talk before the Optical Society of America, presented a paper at the International Conference on Hot Electrons in Semiconductors and served as conference vice-chairman.

1 September 1977 - 31 August 1978. To appreciate our work during this period, the reader has to understand the state of our knowledge concerning the picosecond excite and probe response of germanium at the end of the summer of 1977. We had previously measured the *enhanced* transmission of single optical pulses at 1.06 μm through germanium as a function of incident pulse energy, and we had measured the temporal evolution of this enhanced transmission on a picosecond time scale using the 1.06-μm-excite and 1.06-μm-probe technique. The latter measurements reveal that the probe transmission increases for approximately 100 psec following excitation for a sample temperature of 100 K; however, the rise in probe transmission is less than 40 psec at 300 K. We had initially[5,7] attributed the slower rise in probe transmission at 100 K

to a cooling of a hot electron-hole plasma created by the excitation pulse. Thus, the rise in probe transmission measures the energy relaxation rate and should be a sensitive measure of the electron-phonon coupling constants. The investigations that we reported and discussed previously (see Latham et al.[9]) had subsequently shown that uncertainties in the electron-phonon coupling constants cast doubt on this interpretation. In contrast, Auston et al.[12] had stated that they expected the energy relaxation rate to be too short to account for this rise in probe transmission. They had also suggested[12] that processes omitted from our model, specifically intervalence-band and Coulomb-assisted indirect absorption, might be important. Indeed, they suggested[12] that these processes might introduce a minimum in the absorption versus carrier density curve and, through a private communication, explained that this curve might be combined with Auger recombination to account for the rise in probe transmission. Thus, there were at least three possible explanations for the rise in probe transmission:

(1) the rise, like the "correlation" spike, is an artifact of the experimental technique (an integration effect)

(2) the rise is caused by a cooling of a dense "hot" carrier distribution

(3) the rise is due to Auger recombination combined with an absorption vs. density curve containing a minimum, caused by intervalence-band and Coulomb-assisted indirect absorption.

This was a confusing state of affairs to say the least. Thus, we decided to test the new model suggested by Shank, Auston, McAfee and others and to determine whether or not the rise in probe transmission contained any physics, or whether it was an artifact of the experimental technique. Our goals in these investigations were twofold: (1) to determine the importance

of intervalence-band and Coulomb-assisted indirect absorption events at these carrier densities and (2) if they are important, to determine whether or not they can account for the rise in probe transmission. In order to test these models and these processes, we first needed a probe that was sensitive to Auger recombination as well as indirect and intervalence-band absorption while insensitive to other processes. In particular, the probe should be decoupled from direct absorption processes--note that we attributed the probe rise to a saturation of the direct absorption coefficient as the carriers cooled. Thus, we chose to employ a 1.55 μm (0.8 ev) probe pulse generated by stimulated Raman scattering in Benezene. (The generation of a quality probe pulse by Raman scattering is a challenging problem in itself.) This probe measured the combined free-carrier, intervalence-band, and indirect absorbance, but its quanta were not energetic enough to excite direct band-to-band transitions. We repeated our 1.06 μm excite and probe experiments employing this second probe at 1.55 μm. These studies revealed no minimum in the absorption versus carrier density relationship, and, thus, this relationship cannot be combined with carrier recombination to account for the rise in probe transmission. The contributions of free-carrier, intervalence-band, and Coulomb-assisted indirect absorption, as well as Auger recombination, are significant, and they must be included in any model. In addition, we demonstrated that the rise in probe transmission at 100 K is a real effect and not an artifact of the experimental method. The details of these measurements are contained in Smirl et al.[13, 17] (A preprint is provided in Appendix D).

Finally, Elci[19] conducted theoretical investigations that predicted a great enhancement of the electron-hole recombination in narrow bandgap semiconductors when the gap, or carrier concentration, is adjusted such

that the band-gap energy is approximately equal to the plasmon energy. We are pleased to say that these predictions have been confirmed by measurements reported by Dornhaus and Nimtz at the International Conference on Recombination in Semiconductors, 30 August 1978, Southhampton, U.K.

We have published four papers[13,15,17,19] as a result of our work this year (a total of 9 for the two year period). And we have presented a paper at the Conference on Picosecond Phenomena. In addition, our results have been the subject of theoretical papers by Ferry[14] at Colorado State and by van Driel[15] at the University of Toronto.

In the next section, we detail the direction we expect to take in these studies for the next year.

IV. FUTURE STUDIES

As we have detailed in the previous sections, the number of processes that may be active during the picosecond optical excitation of germanium is staggering. In fact, we listed 16 of these processes or effects in Section II. Many of these processes are important only at extremely high densities. Thus, they are not well studied or understood. As a result, interpretation of initial data is next to impossible. Last year we began an attempt at sorting out the effects of various processes. We shall continue this effort during the next twelve months.

Initially, we shall investigate the origins of the so-called "correlation spike" observed in the probe transmission in germanium. This sharp spike in probe transmission near zero delay in excite and probe experiments at 1.06 μm has been interpreted by Shank and Auston[3] as a scattering of the excitation beam into the probe beam by an index grating formed by the interference of the two beams. As such, the spike in probe transmission should be a correlation of excite pulse and probe pulse. However, other interpretations have been proposed. Ferry[14] has attributed this spike to the formation of a δ-function-like spike in the carrier distribution function and to band narrowing. The origins of this spike will be initially investigated by repeating our original excite-probe experiments, where both excitation and probe pulse have a wavelength of 1.06 μm. However, in these measurements, the probe pulse polarization will be chosen perpendicular to that of the excitation pulse. If the spike is caused by the interference of the probe and excitation beams, the spike should disappear. If the spike remains during the polarization experiments,

then measurements must be performed to determine whether or not study of this spike will provide information on the nonequilibrium distribution function. Supposing, for the moment, that the spike is caused by the interference of the probe and excitation beams, we expect our initial polarization studies to be followed by a detailed study of the surface reflectivity at various angles of incidence (both in the presence and the absence of diffraction effects) in an effort to measure the changes in the real and imaginary part of the index of refraction of germanium under optical excitation. These measurements could provide information concerning carrier number and temperature. A third set of measurements will be performed, assuming diffraction effects are dominant. Two pulses, simultaneously incident on the germanium surface, will form an index grating; the lifetime of this grating will be measured by monitoring the diffracted light from a third, delayed pulse. This third measurement should provide a separation of recombination and diffusion effects. (As the reader may recall from Section II, the original measurements[2] of diffusion effects at these carrier densities were complicated by the reduction in carrier density due to Auger recombination.) In summary, these investigations should provide information concerning the carrier distribution function, real and imaginary part of the index of refraction, and diffusion at large optically-created carrier densities.

In a separate attempt to determine the energy relaxation time of the excited carrier distribution, excite-probe measurements will be performed at various wavelengths. In this application of the excite and probe technique, a high density hot plasma will be created by direct absorption of an excitation pulse with photon energy much greater than the direct band gap in germanium. The evolution of the plasma will

be monitored by a probe pulse with photon energy greater than the band gap but less than the photon energy of the excitation pulse. This second pulse will track the carriers as they cool within the conduction and valence band.

The exact roles of the direct energy band gap and the energy separation between central and indirect conduction band valleys as they relate to band filling and inter-valley scattering will be investigated by repeating many of our previous studies as the band gap of germanium is varied by cycling our sample from 300 K to 14 K in a closed cycle refrigerator. We are also interested in determining whether our approximation of a single termperature for both central and side-valley conduction band electron distributions and valence band hole distributions is a good one.

Finally, we believe one of the most productive experiments might well be to measure the band-to-band photoluminescence spectra on picosecond time scales following optical excitation with a picosecond pulse. It is not clear if the recombination radiation will be intense enough to allow measurement with our present equipment. However, if these measurements are successful, they should provide a temporal indicator of the carrier location within the bands. As such, one should be able to deduce both the distribution function and energy relaxation rates. We expect to observe a vastly accelerated radiative recombination rate from the central valley compared to that expected at lower carrier densities.

The experiments described above are designed to separate and identify the contributions of plasmon emission, phonon-assisted carrier relaxation, diffusion, radiative recombination and other electronic interactions to the ultrafast temporal response of free carrier distributions in semiconductors under intense optical excitation. The assumptions that the

carrier distributions are Fermi-like (and, indeed, spatially independent for thin samples) and that phonon distributions are Bose-like will be tested in these experiments.

V. PROFESSIONAL PUBLICATIONS AND ACTIVITIES

FOR THE PERIOD

1 Sept. 1976 to 31 Aug. 1978

Arthur L. Smirl - Principal Investigator

Publications

1. "Ultrafast Transient Response of Solid State Plasmas: I Germanium: Theory and Experiment," Ahmet Elci, M. O. Scully, A. L. Smirl, and J. C. Matter, Phys. Rev. B 16, 191 (1977).

2. "Pulsewidth Dependence of the Transmission of Ultrashort Optical Pulses in Germanium," John S. Bessey, Bruno Bosacchi, Henry M. van Driel, and Arthur L. Smirl, Phys. Rev. B 17, 159 (1978).

3. "The Role of Phonons and Plasmons in Describing the Pulsewidth Dependence of the Transmission of Ultrashort Optical Pulses through Germanium," W. P. Latham, Jr., A. L. Smirl, A. Elci, and J. S. Bessey, Solid-State Electron. 21, 159 (1978).

4. "Physics of Ultrafast Phenomena in Solid State Plasmas," A. Elci, A. L. Smirl, C. Y. Leung, and M. O. Scully, Solid-State Electron. 21, 151 (1978).

5. "Gauge Invariant Perturbation Theory for the Interaction of Radiation and Matter," Donald H. Kobe and Arthur L. Smirl, Am. J. Phys. 46, 624 (1978).

6. "Simple Laser Pulse Energy Monitor," A. L. Smirl, R. L. Shoemaker, J. B. Hambenne, and J. C. Matter, Rev. Sci. Instrum. 49, 672 (1978)

7. "Picosecond Optical Measurement of Free-Electron, Free-Hole, and Indirect Absorption in Germanium at High Optically-Created Carrier Densities," Arthur L. Smirl, J. Ryan Lindle, and Steven C. Moss, Accepted by Phys. Rev. B for publication November, 1978.

8. "Picosecond Optical Absorption at 1.06 μm and 1.55 μm in Thin Germanium Samples at High Optically-Created Carrier Densities," Arthur L. Smirl, J. Ryan Lindle, and Steven C. Moss, Proceedings of the Conference on Picosecond Phenomena, Springer-Verlag (in press).

9. "Electron-Hole Recombination via Plasmon Emission in Narrow-Gap Semiconductors," Phys. Rev. B 16, 5443 (1977).

Papers

1. "Ultrafast Transient Response of Optically Excited Plasmas in Germanium," Invited paper presented at 1976 Annual Meeting of Optical Society of America, J. Opt. Soc. Am. 66, 1082 (1976).

2. "Gauge Invariant Formulation of the Interaction of Radiation and Matter," D. H. Kobe and A. L. Smirl, AAPT Summer Meeting, 1977, AAPT Announcer 7, 70 (1977).

3. "The Role of Phonons and Plasmons in Describing the Pulsewidth Dependence of the Transmission of Ultrashort Optical Pulses through Germanium," A. L. Smirl, W. P. Latham, A. Elci, and J. S. Bessey, International Conference on Hot Electrons in Semiconductors, Bull. Am. Phys. Soc. 22, 706 (1977).

4. "Picosecond Optical Absorption at 1.06 μm and 1.55 μm in Thin Germanium Samples at High Optically-Created Carrier Densities," A. L. Smirl, J. R. Lindle, and S. C. Moss, Conference on Picosecond Phenomena, May 25, 1978, Hilton Head, S.C.

Other Talks

1. "Ultrashort Optical Pulse Measurement at 10.6 Microns," Larry Tipton, A. L. Smirl, and D. G. Seiler, Texas Academy of Science Meeting at Baylor University, March 10, 1977.

2. "Ultrafast Transient Response of Hot Carriers in Germanium," S. C. Moss, J. R. Lindle, A. L. Smirl, Texas Academy of Science Meeting at Baylor University, March 10, 1977.

3. "Gauge Transformations and Perturbation Theory," D. H. Kobe and A. L. Smirl, Texas Academy of Science Meeting at Baylor University, March 10, 1977.

4. "A Perturbing Question: How Do You Treat the Interaction of Electromagnetic Radiation and Matter?", D. H. Kobe and A. L. Smirl, Texas Section of AAPT Meeting at Southern Methodist University, November 5, 1977.

5. "A Simple, Accurate, Inexpensive Energy Monitor for Short Laser Pulses," J. R. Lindle, S. C. Moss, and A. L. Smirl, Texas Academy of Science Meeting at Texas Tech, March 9, 1978.

6. "Measurement of the Ultrafast Relaxation of Saturation Phenomena in Semiconductors," S. C. Moss, J. R. Lindle, and A. L. Smirl, Texas Academy of Science Meeting at Texas Tech, March 9, 1978.

7. "Piezo-Transmission of p-type Germanium at CO_2 Laser Wavelengths," L. Tipton, D. G. Seiler, and A. L. Smirl, Texas Academy of Science Meeting at Texas Tech, March 9, 1978.

VI. VITA

SMIRL, ARTHUR LEE

Assistant Professor Physics, North Texas State University

Personal

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Education

Ph.D.	Optical Sciences	University of Arizona	1975
M.S.E.	Electrical Engineering	University of Michigan	1969
B.S.	Electrical Engineering	Lamar University	1968
B.S.	Mathematics	Lamar University	1968

Experience

1975 - Assistant Professor, North Texas State University, Denton Texas
1972-75 - Research Associate, University of Arizona, Tucson, Arizona
1968-71 - Staff member, Sandia Laboratories, Albuquerque, New Mexico

Research Specialization

Optical Interactions in Semiconductors
Picosecond Spectroscopy
Laser Physics

Publications - A. L. Smirl

1. "Holographic Recording with Limited Laser Light," C. D. Leonard and A. L. Smirl, Appl. Opt. 10, 625 (1971).

2. "Nonlinear Absorption and Ultrashort Carrier Relaxation Times in Germanium Under Irradiation by Picosecond Pulses," Chandler J. Kennedy, John C. Matter, Arthur L. Smirl, Hugo Weichel, Frederic A. Hopf, and Sastry V. Pappu, Phys. Rev. Lett. 32, 419 (1974).

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* Supported in part by the Office of Naval Research

Papers - A. L. Smirl

1. "Available Light Holography," A. L. Smirl and C. D. Leonard, 1969 Annual Meeting of the Optical Society of American, J. Opt. Soc. Am. 59, 1530 (1969).

'2. "Ultrafast Transient Response of Optically Excited Plasmas in Germanium," Invited paper presented at 1976 Annual Meeting of Optical Society of America, J. Opt. Soc. Am. 66, 1082 (1976).

*3. "Gauge Invariant Formulation of the Interaction of Radiation and Matter," D. H. Kobe and A. L. Smirl, AAPT Summer Meeting, 1977, AAPT Announcer 7, 70 (1977).

*4. "The Role of Phonons and Plasmons in Describing the Pulsewidth Dependence of the Transmission of Ultrashort Optical Pulses through Germanium," A. L. Smirl, W. P. Latham, A. Elci, and J. S. Bessey, International Conference on Hot Electrons in Semiconductors, Bull. Am. Phys. Soc. 22, 706 (1977).

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Other Presentations Given at

Scientific Meetings - A. L. Smirl

*1. "Ultrashort Optical Pulse Measurement at 10.6 Microns," Larry Tipton, A. L. Smirl, and D. G. Seiler, Texas Academy of Science Meeting at Baylor University, March 10, 1977.

*2. "Ultrafast Transient Response of Hot carriers in Germanium," S. C. Moss, J. R. Lindle, A. L. Smirl, Texas Academy of Science Meeting at Baylor University, March 10, 1977.

*3. "Gauge Transformations and Perturbation Theory," D. H. Kobe and A. L. Smirl, Texas Academy of Science Meeting at Baylor University, March 10, 1977.

*4. "A Perturbing Question: How Do You Treat the Interaction of Electromagnetic Radiation and Matter?", D. H. Kobe and A. L. Smirl, Texas Section of AAPT Meeting at Southern Methodist University, November 5, 1977.

*5. "A Simple, Accurate, Inexpensive Energy Monitor for Short Laser Pulses," J. R. Lindle, S. C. Moss, and A. L. Smirl, Texas Academy of Science Meeting at Texas Tech, March 9, 1978.

*6. "Measurement of the Ultrafast Relaxation of Saturation Phenomena in Semiconductors," S. C. Moss, J. R. Lindle, and A. L. Smirl, Texas Academy of Science Meeting at Texas Tech, March 9, 1978.

*7. "Piezo-Transmission of p-type Germanium at CO_2 Laser Wavelengths," L. Tipton, D. G. Seiler, and A. L. Smirl, Texas Academy of Science Meeting at Texas Tech, March 9, 1978.

* Supported in part by the Office of Naval Research

VII. BUDGET SUMMARY

1 Sept. 1976 to 31 Aug. 1978

	Actual
A. Salaries and Wages:	
1. Principal Investigator	$14,044.43
2. Research Associate	25,911.03
3. Research Assistants	10,199.79
4. Secretary	856.00
Total Salaries	$51,011.25
B. Benefits:	5,611.51
C. Total Salaries and Benefits:	56,622.76
D. Permanent Equipment:	7,237.59
E. Expendable Supplies and Equipment:	14,820.46
F. Travel:	2,801.69
G. Publications Cost:	3,517.50
H. Total Direct Cost:	$85,000.00
I. Indirect Costs:	8,000.00
J. Total Costs:	$93,000.00

Note: The original contract was for $80,000 over a period of two years. At the end of the first year $13,000 was added to the salaries category.

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APPENDIX A

SIMPLE LASER PULSE MONITOR

Simple laser pulse energy monitor

Arthur L. Smirl

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R. L. Shoemaker, J. B. Hambenne,[a] and J. C. Matter[b]

Optical Sciences Center, University of Arizona, Tucson, Arizona 85721

(Received 28 September 1977)

A simple and inexpensive relative energy monitor for short laser pulses is described. The three basic units of this system are an integrating detector circuit, an amplifier, and a peak detector and hold circuit. With this device one can measure optical pulse energies (at 1.06 μm) as small as 1.0 nJ with an electronic accuracy of $\sim 1\%$.

In pulsed laser experiments, one often wants to measure quantities such as the shot-to-shot variation in laser pulse energy or the fraction of laser power reflected from or transmitted through a sample.[1,2] Such measurements require only a determination of the relative energy in a laser pulse and can easily be made by using a reverse-biased photodiode as the laser pulse detector and charging a capacitor with the photodiode output. The problem then reduces to measuring the charge on the capacitor. The usual way of doing this is to allow the capacitor to discharge through the 1-MΩ input impedance of an oscilloscope and to photograph the resulting RC decay curve. Unfortunately, this technique has rather limited accuracy and the cost of photographic film can become prohibitive if many measurements are to be made.

A much better method is to use a sample-and-hold circuit so that the capacitor charge can be read out as a dc voltage. A sample-and-hold device with a tracking bandwidth on the order of 1 MHz and a memory decay rate of ~ 0.5 mV/s is needed for this approach, however, and integrated circuit or hybrid devices meeting these specifications cannot be obtained commercially. In this note we present a simple, inexpensive circuit which holds the capacitor charge for about 30 s, allowing one to easily measure the relative pulse energy with any dc voltmeter. The electronic accuracy of the circuit is about 1%.

The circuit is shown in Fig. 1 and consists of three basic units: an integrating detector, an amplifier, and a peak detector-and-hold circuit. The detector circuit is fairly conventional and uses a United Detector Technology PIN 6D silicon photodiode to charge a 100-pF capacitor. This is followed by a 10× gain noninverting amplifier. The peak detector-and-hold circuit uses two operational amplifiers. The first is effectively an inverting, unity gain amplifier which charges the 0.01-μF track and hold capacitor C_2. Both this and the previous amplifier must be wideband (settling time

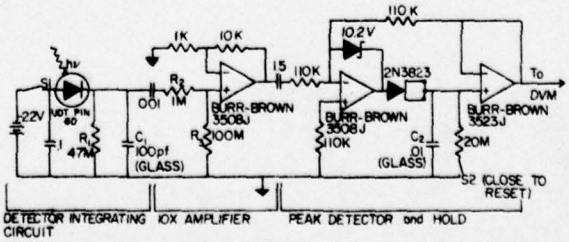

FIG. 1. Energy monitor for short pulse lasers. (Resistance is in ohms and capacitance is in μF, unless otherwise noted.)

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<50 μs) in order to accurately capture the input waveform. The offset voltage of the unity gain amplifier is important and may be conveniently checked by shunting the FET by a resistor (e.g., 1 kΩ) and monitoring the output of the buffer amplifier. The second op-amp in the peak detect-and-hold circuit is a high input impedance, ultralow bias current, unity gain buffer amplifier which allows the voltage across capacitor C_2 to be read while drawing negligible charge from it. A 2N3823 FET is used as a high reverse impedance diode to clamp the capacitor voltage at the peak of the input waveform. Care should be exercised in choosing capacitor C_2, since many capacitors were found to have a leakage resistance that was much less than the back resistance of the FET or the input impedance of the buffer op amp. A glass capacitor proved adequate for our purposes.

The unique feature of our circuit is the use of an inexpensive ultralow input offset current op-amp (Burr–Brown 3523J) having limited bandwidth as the buffer amplifier. The limited bandwidth causes a brief period of linear charging with no feedback during which time the tracking capacitor is not following the input signal. Thus for small signals applied to the peak detector and hold, the voltage on the tracking capacitor will actually overshoot the peak of the input. In our circuit this restricts the dynamic range of the input to 0.6–10 V, rather than the 0–10 V one would have in an ideal buffer amplifier.

This sacrifice of dynamic range allows construction of a rather inexpensive peak detector and hold with very high performance. All the circuit components for our energy monitor cost less than $75, including the photodiode (but not a digital voltmeter).

A simple method of dynamically testing the accuracy and linearity of the peak detector is to apply a repetitive pulse train as an input while the track-and-hold capacitor C_2 is shunted by a resistor to provide an *RC* time constant less than the repetition rate. The waveforms may then be observed on an oscilloscope. To realize the full potential of this circuit, it should be mounted in a well shielded copper box, and the detector should be optically baffled to eliminate stray light.

The support of the National Science Foundation and the Office of Naval Research is gratefully acknowledged.

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[b] Present address: Sandia Laboratories, Projects Division 1245, Albuquerque, NM 87115.

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APPENDIX B

ELECTRON-HOLE RECOMBINATION VIA PLASMON EMISSION IN NARROW-GAP SEMICONDUCTORS

PHYSICAL REVIEW B VOLUME 16, NUMBER 12 15 DECEMBER 1977

Electron-hole recombination via plasmon emission in narrow-gap semiconductors

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(Received 16 August 1977)

When the plasma frequency coincides with the band gap in a narrow-gap semiconductor like $(Pb_{1-x}Sn_x)Se$, the electron-hole recombination is quite fast, in the picosecond range. Plasmon lifetime is calculated and taken into account in the calculation of the recombination rate. Experiments in which the plasmon-assisted recombination may be observed directly are proposed.

I. INTRODUCTION

Recent picosecond optical pulse measurements on Ge have shown that plasmons play a significant role in the nonlinear transmission of intense optical pulses. The optical properties of a Ge wafer irradiated by an intense Nd:glass laser pulse are significantly altered by the rapid recombinations of electrons and holes via plasmon emissions at high plasma densities. Due to these recombinations, nonlinear light absorption in Ge is observed to decrease relatively slowly and saturate at a higher absorption level as optical pulse energy is increased than otherwise would be expected. The dense plasmas created by the picosecond optical pulses display other properties that are also attributed to the plasmon-assisted electron-hole recombination.[1,2]

However, in all these measurements this type of recombination is observed only *indirectly*, since other equally rapid processes are taking place simultaneously while the energetic optical pulse is being absorbed in Ge. It would be desirable to observe the electron-hole recombination via plasmon emission more directly. The best materials in which this recombination can be more directly investigated are the narrow-gap semiconductor alloys $(Pb_{1-x}Sn_x)Se$, $(Pb_{1-x}Sn_x)Te$, and $(Cd_{1-x}Hg)Te$. In these materials, the energy gap E_G between conduction and valence bands is direct and can be continuously varied through zero by changing alloy composition or by applying hydrostatic pressure or magnetic field.[3-7] Some time ago, Wolff[8,9] suggested that it may be possible to induce plasma-wave instability in these materials by adjusting E_G to coincide with the plasma frequency $\hbar\omega_p$, then pumping holes or electrons into the semiconductor by an electron or laser beam. Whether or not stimulated plasmon emission occurs when electrons or holes are externally pumped, *there is an enhancement of electron-hole recombination if* $E_G \sim \hbar\omega_p$. If $E_G \sim \hbar\omega_p$, the pumped electrons and holes recombine via plasmon emission at a rate two to three orders of magnitude larger than the rate at which they recombine via photon emission.[10] Plasmons which are emitted by the recombining electron-hole pairs generally have long wavelength (~ approximately sample dimension).

To illustrate the physics of the situation, let us consider a specific semiconductor $(Pb_{1-x}Sn_x)Se$. Its band structure is schematically shown in Fig. 1. The band gap is at L, and there are four conduction-band valleys.[11] Let us assume that temperature is low and that the conduction band is filled up to the Fermi level E_F which is considerably larger than E_G. A typical conduction-band electron density is $n_c \sim 10^{18}$ cm^{-3}. Now let us suppose that the semiconductor is pumped either optically (quanta of energy $h\nu_L$) or with an electron beam and a small density of holes (10^{14}–10^{15} cm^{-3}) is created. The thermalization rate for holes is relatively short[9] ($\lesssim 10^{-13}$ sec). Therefore holes rapidly lose energy and collect near the band edge as shown in Fig. 2. The hole Fermi level δE_p is assumed to be small; $\delta E_p \ll E_G$. Holes and electrons which are near the band edge begin to recombine. This recombination can occur through many mechanisms, which are discussed in detail in a review paper by Bonch-Bruevich and

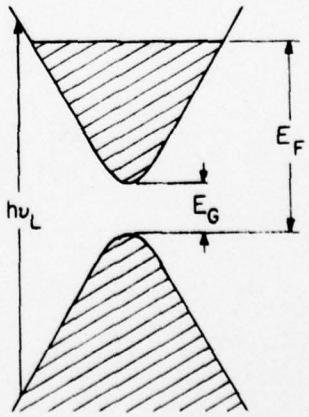

FIG. 1. $(Pb_{1-x}Sn_x)$ Se band structure.

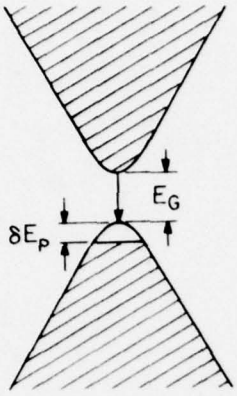

FIG. 2. Thermalized hole distribution.

Landsberg.[10] The special feature of the semiconductors mentioned above is that when $E_G \sim \hbar\omega_p$, plasmon emission is exceedingly rapid and electron-hole recombination is therefore essentially determined by plasmon mechanism alone. It is the long-range portion of the electronic Coulomb field which induces the rapid recombinations in these materials.

In Wolff's original work on the plasma-wave instability,[9] the plasmon lifetime was taken as infinite in his calculation of the spontaneous plasmon-assisted recombination rate. Therefore the rate he obtained goes to zero for $\hbar\omega_p \leq E_G$. However plasmon decay in the semiconductor alloys mentioned above is rapid even for plasmons which have long wavelengths. The enhanced recombination can actually occur for band gaps larger than $\hbar\omega_p$ as a result of the short lifetime of plasmons and the consequent broadening of their energy.

In this paper, the spontaneous recombination rate of holes in an n-type narrow-gap semiconductor is calculated for finite plasmon lifetime. The plasmon lifetime is calculated in Sec. III, and found to be about 1 psec. The calculated recombination rate turns out to be quite large for a relatively wide interval of band gap values (see Fig. 5). Although the plasmon-assisted recombination rate is somewhat reduced for $E_G < \hbar\omega_p$ compared to that for $E_G < \hbar\omega_p$, it is still considerably larger than the rates for other recombination mechanisms. For example, it is about two orders of magnitude larger than the rate of optical recombinations. In Sec. IV, experiments in which the plasmon-assisted recombination can be directly observed are suggested and discussed in some detail.

II. SPONTANEOUS RECOMBINATION RATE OF HOLES

In this section, the spontaneous recombination rate of holes is calculated for the carrier distributions illustrated in Fig. 2. In the following calculation we make use of the propagator for plasmons and the self-energy method,[12-14] which are well known and will not be elaborated upon here. Let us note that the plasmon lifetime, or more precisely, the width of the plasmon resonance, which is $\hbar \times$ (plasmon lifetime)$^{-1}$, enters into the recombination rate calculation quite naturally when the self-energy method is used. In this section the plasmon resonance width is treated as an unknown parameter. It is evaluated in Sec. III.

Figure 3 shows the diagram for the recombination via plasmon emission. For a hole of wave vector $\vec{k}$, the recombination rate is given by[9]

$$\frac{1}{\tau_R(\vec{k})} = 2\int \frac{d\vec{q}}{(2\pi)^3}\left[\frac{4\pi e^2\hbar}{q^2\epsilon_\infty}\right]\left[\frac{\vec{q}\cdot\vec{P}_{cv}}{mE_G}\right]^2 \times \mathrm{Im}\left[\frac{1}{\epsilon(\vec{q}, E_c(\vec{k}+\vec{q}) - E_v(\vec{k}))}\right] \quad (1)$$

Here m is the bare electron mass; $\vec{P}_{cv}$ is the interband matrix element of the momentum operator; and E_c and E_v are electron and hole energies, respectively. ϵ_∞ is the high frequency dielectric constant, and $\epsilon_\infty\epsilon(\vec{q}, \omega)$ is the overall dielectric function.[8] Note that

$$\lim_{\omega\to\infty} \epsilon(q, \omega) = 1 . \quad (2)$$

The $\vec{P}_{cv}$ factor in Eq. (1) comes from the interband matrix element of the density fluctuation operator, which, for long-wavelength fluctuations, is given by[15]

$$\langle c\vec{k}|e^{i\vec{q}\cdot\vec{r}}|v\vec{k}'\rangle \simeq -(\vec{q}\cdot\vec{P}_{cv}/mE_G)\delta^3(\vec{k}'+\vec{q}-\vec{k}) . \quad (3)$$

For the energy bands of $(Pb_{1-x}Sn_x)Se$, one can use the two-band model.[16] Then

$$E_c(\vec{k}) = \frac{E_G}{2} + \left[\left(\frac{E_G}{2}\right)^2 + E_G\frac{\hbar^2\vec{k}^2}{2m_{BE}}\right]^{1/2} , \quad (4a)$$

$$E_v(\vec{k}) = \frac{E_G}{2} - \left[\left(\frac{E_G}{2}\right)^2 + E_G\frac{\hbar^2\vec{k}^2}{2m_{BE}}\right]^{1/2} . \quad (4b)$$

m_{BE} is the effective mass at the band edge

$$\frac{1}{m_{BE}} = \left(\frac{1}{\hbar^2 k}\frac{dE_c(k)}{dk}\right)_{k=0} . \quad (5)$$

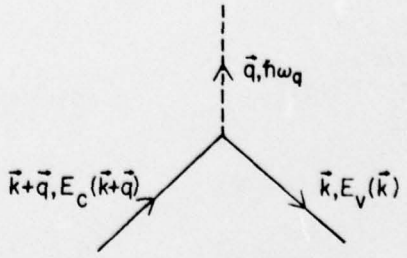

FIG. 3. Plasmon emission.

It is related to $\vec{P}_{cv}$ by the equation

$$1/m_{BE} = 2|\vec{P}_{cv}|^2/3m^2E_G . \quad (6)$$

Near the plasmon pole, Eq. (1) becomes

$$\frac{1}{\tau_R(\vec{k})} = \frac{e^2\hbar^2}{4\pi^2\epsilon_\infty} \times \int d\vec{q} \left| \frac{\hat{q}\cdot\vec{P}_{cv}}{mE_G} \right|^2 \times \omega_q \left(\frac{\Delta(\vec{q})}{[\hbar\omega_q - E_c(\vec{k}+\vec{q}) + E_v(\vec{k})]^2 + [\frac{1}{2}\Delta(\vec{q})]^2} \right) , \quad (7)$$

where $\hbar\Delta(\vec{q})^{-1}$ is the lifetime and ω_q is the frequency for a plasmon of wave vector $\vec{q}$. It is clear from (7) that when $E_G \sim \hbar\omega_p$, where ω_p is the plasma frequency for $q=0$, the resonance is set at $k\sim 0$ and $q\sim 0$. The hole densities we have in mind are small, and holes are confined to a small region near the band edge. Let us therefore set $\vec{k}=0$. Near the pole, $\Delta(\vec{q})$ can be approximated by $\Delta(0)$ and thus treated as constant. We will also neglect plasmon dispersion and set $\omega_q = \omega_p$. ω_p is given by

$$\omega_p = (4\pi e^2 n_c/\epsilon_\infty m_{1S})^{1/2} , \quad (8)$$

where n_c is the density of electrons in the conduction band and m_{1S} is the effective mass at the Fermi surface

$$\frac{1}{m_{1S}} = \left(\frac{1}{\hbar^2 k} \frac{dE_c(k)}{dk} \right)_{k=k_F} . \quad (9)$$

The hole-density contribution to the plasma frequency is neglected since the hole density is assumed to be small ($\sim 10^{14}$–10^{15} cm^{-3}, compared to $n_c \sim 10^{18}$ cm^{-3}).

Finally, for small $\vec{q}$, we can approximate (4a) by

$$E_c(\vec{q}) = E_G + \hbar^2\vec{q}^2/2m_{BE} . \quad (10)$$

Thus the recombination rate in Eq. (7) becomes, at $\vec{k}=0$,

$$\frac{1}{\tau_R(0)} = \frac{\hbar^2 e^2\omega_p}{4\pi^2\epsilon_\infty} \times \int d\vec{q} \left(\frac{\hat{q}\cdot\vec{P}_{cv}}{mE_G} \right)^2 \times \frac{\Delta(0)}{(\hbar\omega_p - E_G - \hbar^2q^2/2m_{BE})^2 + [\frac{1}{2}\Delta(0)]^2} . \quad (11)$$

This integral can be evaluated analytically. If we define

$$\rho \equiv E_G/\hbar\omega_p \quad (12)$$

and

$$\zeta \equiv \Delta(0)/2\hbar\omega_p , \quad (13)$$

then Eq. (11) becomes

$$\frac{1}{\tau_R(0)} = \left(\frac{6^{1/2}e^2\omega_p m\zeta}{\hbar\epsilon_\infty|\vec{P}_{cv}|} \right) \times \{\rho[(\rho-1)^2+\zeta^2]^{1/2} + \rho(\rho-1)\}^{-1/2} . \quad (14)$$

Note that as the plasmon width $\Delta(0)$ goes to zero,

$$\lim_{\zeta\to 0} \zeta\{\rho[(\rho-1)^2+\zeta^2]^{1/2} + \rho(\rho-1)\}^{-1/2} = \begin{cases} 0 & \text{for } \rho > 1 , \\ 2^{1/2}\rho^{-1/2}(1-\rho)^{1/2} & \text{for } \rho < 1, \end{cases}$$

and (14) reduces to the expression given by Wolff[9] (neglecting δE_p, the hole Fermi energy).

III. PLASMON LIFETIME

In order to complete the calculation of the spontaneous recombination rate, we need the plasmon resonance width for long-wavelength plasmons, i.e., $\Delta(0)$. Here, we calculate the plasmon decay for the lowest-order perturbation diagrams, since higher-order diagrams are not expected to contribute to the decay in any significant amount. We have only indicated the outlines of the calculation in the text. The details are discussed in the Appendix.

$(Pb_{1-x}Sn_x)Se$, $(Pb_{1-x}Sn_x)Te$, and $(Cd_{1-x}Hg_x)Te$ are relatively degenerate when $\hbar\omega_p \sim E_G$, for both n-type and p-type samples. Long-wavelength plasmons decay primarily via impurity scattering in these materials. As the plasmon wavevector $q\to 0$, other plasmon decay mechanisms—e.g., Landau damping and phonon scattering—are rela-

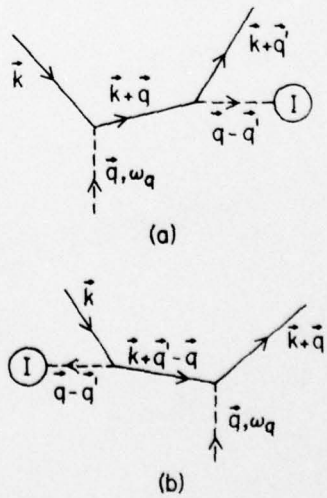

FIG. 4. Plasmon-impurity scattering.

tively unimportant,[17] and will be ignored in this paper.

The lowest order plasmon-impurity scattering is shown in Figs. 4(a) and 4(b). Here a plasmon breaks up into an electron and a hole in the fixed field of the impurity. The impurity imparts enough momentum to the incoming plasmon to permit the transfer of the plasmon energy to individual particle states, i.e., to raising an electron above the Fermi level, leaving an empty state behind, in the conduction band. Since the impurity is fixed in space, no energy is given to or taken from it. As the initial plasmon momentum goes to zero, the contribution of these diagrams remains constant and large.

The net amplitude for the plasmon decay in a screened impurity field is the sum of the two diagrams shown in Fig. 4 (Ref. 18):

$$i\left(\frac{4\pi e^2\hbar^2\omega_q}{q^2\epsilon_\infty}\right)^{1/2}\left(\frac{4\pi e^2}{\epsilon_\infty}\right)\left(\frac{1}{|\vec{q}-\vec{q}'|^2+\kappa^2}\right)\sum_{i=1}^{n_i} e^{-i(\vec{q}-\vec{q}')\cdot\vec{R}_i}\left(\frac{1}{\hbar\omega_q - E_c(\vec{k}+\vec{q})+E_c(\vec{k})}\right.$$

$$\left.-\frac{1}{\hbar\omega_q - E_c(\vec{k}+\vec{q}')+E_c(\vec{k}+\vec{q}'-\vec{q})}\right),$$

where $\vec{k}$, $\vec{q}$, and $\vec{q}'$ are as defined in the diagrams, R_i refers to the position of an impurity, n_i is the number of impurities in the volume (which is taken to be unity in the calculation), and κ is the Fermi-Thomas wave vector

$$\kappa^2 = 3\omega_p^2/v_F^2 = 3\omega_p^2 m_{FS}^2/\hbar^2 k_F^2 . \tag{15}$$

The absolute square of this amplitude contains the factor

$$\sum_{i=1}^{n_i}\sum_{j=1}^{n_i} e^{-i(\vec{q}-\vec{q}')\cdot(\vec{R}_i-\vec{R}_j)} .$$

Impurities are distributed randomly and therefore to a good approximation we may take the sum of the diagonal terms ($i=j$) alone,[19] which gives n_i. Let us define the partial width

$$\Gamma(\vec{q},\vec{q}',\vec{k}) = \left(\frac{2\pi}{\hbar}\right)n_i\left(\frac{2\pi e^2\hbar^2\omega_q}{q^2\epsilon_\infty}\right)\left(\frac{4\pi e^2}{\epsilon_\infty}\right)^2\left(\frac{1}{|\vec{q}-\vec{q}'|^2+\kappa^2}\right)^2$$

$$\times\left(\frac{1}{\hbar\omega_q - E_c(\vec{k}+\vec{q})+E_c(\vec{k})} - \frac{1}{\hbar\omega_q - E_c(\vec{k}+\vec{q}')+E_c(\vec{k}+\vec{q}'-\vec{q})}\right)^2 \delta(E_c(\vec{k}+\vec{q}')-E_c(\vec{k})+\hbar\omega_q). \tag{16}$$

Integrating over all possible final states with appropriate weighting factors, we then obtain the plasmon resonance width at the plasmon wave vector $\vec{q}$:

$$\Delta(\vec{q}) = \int\frac{2d\vec{k}}{(2\pi)^3}\int\frac{d\vec{q}'}{(2\pi)^3}\,\Gamma(\vec{q},\vec{q}',\vec{k})f_{\vec{k}}^c(1-f_{\vec{k}+\vec{q}'}^c), \tag{17}$$

where $f_{\vec{k}}^c$ is the Fermi function for electrons with a temperature $T=(k_B\beta)^{-1}$,

$$f_{\vec{k}}^c = 1/(e^{\beta[E_c(k)-E_F]}+1) . \tag{18}$$

The integrals in Eq. (17) are evaluated in the Appendix for $q\to 0$ and $T\to 0$. Within the approximations indicated there, Eq. (17) for $\vec{q}=0$ is

$$\Delta(0) \simeq \Delta_0\left\{\frac{32\xi^4+72\xi^2+30-6\lambda^2\xi^2(5+2\xi^2)}{3\xi^2(1+\xi^2)} + (2+\lambda^3)\ln\left(\frac{1+\xi^2(1+\lambda)^2}{1+\xi^2(1-\lambda)^2}\right)+3\lambda^2\ln|1+2\xi^2(\lambda^2+1)+\xi^4(\lambda^2-1)^2|\right.$$

$$\left.-\frac{1}{\xi}\left(\frac{5}{\xi^2}+9\right)[\tan^{-1}(\xi\lambda+\xi)-\tan^{-1}(\xi\lambda-\xi)]\right\}, \tag{19}$$

where the parameters are

$$\Delta_0 \equiv e^4 m^4\omega_p^2 n_i/12\pi e_\infty^2 m_{FS}|\vec{P}_{cv}|^4 n_c , \tag{20}$$

$$\xi \equiv m\hbar k_F/m_{FS}|\vec{P}_{cv}| , \tag{21}$$

and

$$\lambda = (2E_F - E_G)/\hbar\omega_p. \tag{22}$$

To get some feeling for the actual numbers involved in these calculations, let us consider a sample of $(Pb_{1-x}Sn_x)Se$ for which $x \sim 0.18$ and therefore $E_G \sim 0.03$ eV.[5] For these alloys, $\epsilon_\infty \sim 20$. Rabii's measurements on PbSe give[20]

$$\hbar^2 |\vec{P}_{cv}|^2/3m^2 \simeq 1.85 \times 10^{-30} \text{ erg}^2\text{cm}^2. \tag{23}$$

Let $n_c = 10^{18}$ cm^{-3}. In the limit of degenerate carriers

$$k_F = (\tfrac{1}{4}\pi^2 n_c)^{1/3} \simeq 2 \times 10^6 \text{ cm}^{-1}. \tag{24}$$

From (4a) and (6) one finds

$$E_F = \frac{E_G}{2} + \left[\left(\frac{E_G}{2}\right)^2 + \frac{\hbar^2|\vec{P}_{cv}|^2}{3m^2}k_F^2\right]^{1/2}. \tag{25}$$

When $E_G \sim 0.03$ eV, the last term under the radical sign is an order of magnitude larger than the first one.[21] Neglecting the latter, (25) can be approximated by

$$E_F = \frac{E_G}{2} + \frac{\hbar k_F|\vec{P}_{cv}|}{m\sqrt{3}} \simeq \frac{E_G}{2} + 0.058 \text{ eV}. \tag{26}$$

Using (26), one then finds that

$$m_{FS}/m \simeq \sqrt{3}\,\hbar k_F/|\vec{P}_{cv}| \simeq 4.5 \times 10^{-2}, \tag{27a}$$

$$\xi \simeq 1/\sqrt{3}, \tag{27b}$$

$$\hbar\omega_p = \hbar(4\pi e^2 n_c/\epsilon_\infty m_{FS})^{1/2} \simeq 0.034 \text{ eV}, \tag{27c}$$

and

$$\lambda = 2k_F|\vec{P}_{cv}|/\omega_p m\sqrt{3} \simeq 3.0. \tag{27d}$$

Therefore $\Delta(0)$ is independent of E_G as long as (26) is valid. Setting $n_i \simeq n_c$, we find $\Delta_0 \sim 2 \times 10^{-3}$ eV and $\Delta(0) \simeq 10^{-3}$ eV. Thus, the lifetime of long-wavelength plasmons is about 1 psec.

The recombination rate versus $\rho = E_G/\hbar\omega_p$ for $\Delta(0) \simeq 10^{-3}$ eV is plotted in Fig. 5. It is seen that electron-hole recombination is enhanced and extremely rapid even for $E_G > \hbar\omega_p$. It is also seen that the recombination rate varies as $\Delta(0)\rho^{-1/2}(\rho-1)^{-1/2}$ for $\rho > 1$. This portion of the curve arises entirely from the finite plasmon lifetime.

It might be possible to reproduce this theoretical curve experimentally. The rapidity of the recombination might also be of some use in new devices. We now turn our attention to this subject.

IV. EXPERIMENTAL OBSERVATION AND DISCUSSION

For the sake of definiteness, we will continue to refer to a specific n-type $(Pb_{1-x}Sn_x)Se$ sample. However, only a slight modification of the theoretical discussion offered here applies equally well to p-type $(Pb_{1-x}Sn_x)Se$, and to both n- and p-type $(Pb_{1-x}Sn_x)Te$ and $(Cd_{1-x}Hg_x)Te$.

One direct method of studying the enhanced recombination is to do a reflectivity experiment. Optical pulses of extremely short duration (a few picoseconds) can be achieved by mode-locked lasers.[22] These pulses can be used to measure the decay in the density of electrons and holes which are pumped optically. The schematic of such an experiment is shown in Fig. 6. The sample is assumed to be thin, $W \sim 1$ μm. The optical pulse from the second harmonic of a CO_2 laser creates the holes. For the second harmonic of the CO_2 laser, $h\nu_L = 0.234$ eV $> 2E_F - E_G$, and the light absorption coefficient of the semiconductor at this frequency is $\alpha(\nu_L) \sim 10^4$ cm^{-1}. Another pulse, from the first harmonic of the CO_2 laser $[h\nu = 0.117$ eV

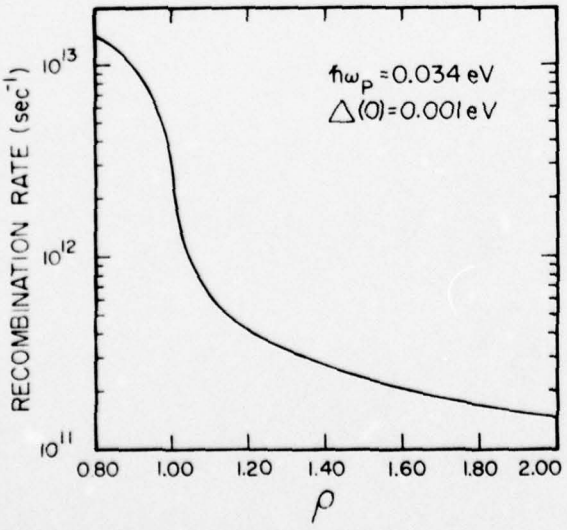

FIG. 5. Plasmon-assisted recombination rate; $\rho \equiv E_G/\hbar\omega_p$.

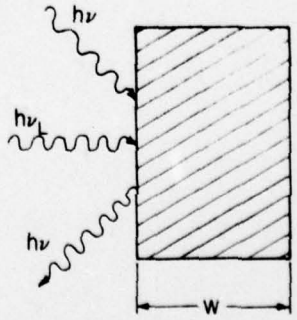

FIG. 6. Schematic of reflectivity experiment.

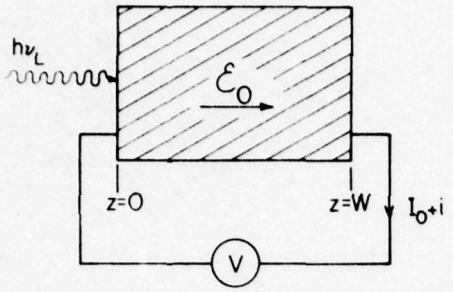

FIG. 7. Schematic of photoconductivity experiment.

$\simeq 2E_F - E_G$; $\alpha(\nu) < \alpha(\nu_L)$], is incident on the sample (normal or at an angle), but is delayed compared to the first pulse. The reflectivity of the second pulse can be measured and related to the hole density created by the first. The time interval between the two pulses can be varied to observe the decrease in the hole density at different delay times. For thin samples ($W \sim \alpha^{-1}$), the decay in the hole density is entirely due to recombinations[23]

$$n_h \propto \exp(-t_{\text{delay}}/\tau_R). \tag{28}$$

Such thin samples can be grown epitaxially. In such an experiment, hydrostatic pressure can be applied to the sample by putting it into a diamond cell. Since E_G varies with hydrostatic pressure ($dE_G/dP = -8.65 \times 10^{-6}$ eV/bar),[24] $1/\tau_R$ can be mapped experimentally as a function of E_G.

The proposed experiment is quite similar in technical detail to those discussed in Refs. 1, 2, and 25. For more information on experimental technique, the reader is referred to these papers and to the references cited therein.

Although the recombination rate is rapid, it is possible to do a photoconductivity experiment to measure it. Since such an experiment allows $1/\tau_R$ to be varied without changing E_G, we will discuss it in some detail. The schematic of the experiment is shown in Fig. 7. A thin wafer of thickness W is subjected to a drift field $\mathcal{E}_0$. There is a current I_0 flowing through the sample. An optical beam, e.g., from a cw CO laser, is turned on and creates holes. Holes thermalize and collect, not, however, at the band edge as discussed previously, but at a region below the band edge because of the drift induced by the constant electric field.[26] This is illustrated in Fig. 8. The shift of the hole distribution from the top of the valence band is, approximately,

$$\Delta E = \tfrac{1}{2} m_H (\mu_h \mathcal{E}_0)^2, \tag{29}$$

where μ_h is the hole mobility. The expression for the recombination rate given in (14) can still be used if E_G in ρ is replaced by an effective gap

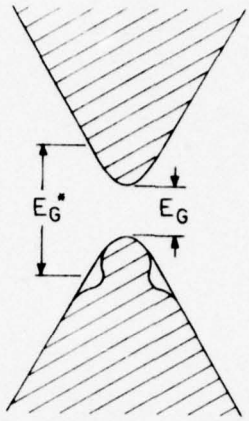

FIG. 8. Hole distribution in constant electric field.

$$E_G^* = E_G + 2\Delta E. \tag{30}$$

Thus the recombination rate is dependent on the electric field applied. The recombination rate can therefore be changed by varying the electric field, as well as by varying the band gap.

When the laser beam is on, holes are generated near $z = 0$. Holes drift and diffuse in the direction of the electric field; many of them recombine. But if the sample thickness is small and the applied electric field large, then there is a sufficient number of holes reaching the surface at $z = W$ to give an appreciable photoinduced current. The photoinduced hole current density is given by

$$j_h = e\mu_h \mathcal{E}_0 n_h - eD_h \frac{dn_h}{dz}, \tag{31}$$

where n_h is the photogenerated hole density and D_h is the hole diffusion constant. In steady state,

$$\frac{1}{e}\frac{d}{dz} j_h + \frac{n_h}{\tau_R} \simeq G\delta(z), \tag{32a}$$

where G = (light speed) × (the number of photons per unit volume in the light beam). Since the photogenerated holes are extracted at $z = W$,

$$n_h(W) = 0. \tag{32b}$$

The solution of (32a) and (32b) is given by

$$n_h(z) = 2G\left[\exp\left(\frac{\mu_h \mathcal{E}_0 z}{2D_h}\right)\right] \sinh\left(\frac{W-z}{l_h}\right) \times \left[\mu_h \mathcal{E}_0 \sinh\left(\frac{W}{l_h}\right) + 2D_h l_h \cosh\left(\frac{W}{l_h}\right)\right]^{-1}, \tag{33a}$$

where

$$l_h \equiv \left[\left(\frac{\mu_h \mathcal{E}_0}{2D_h}\right)^2 + \frac{1}{D_h \tau_R}\right]^{-1/2}. \tag{33b}$$

Thus the current density is at $z = W$,

$$j_h(z=W) = \left(\frac{2eGD_h}{l_h}\right) \exp\left(\frac{\mu_h \mathcal{E}_0 W}{2D_h}\right) \times \left[\mu_h \mathcal{E}_0 \sinh\left(\frac{W}{l_h}\right) + 2D_h l_h \cosh\left(\frac{W}{l_h}\right)\right]^{-1} . \quad (34)$$

For $(Pb_{1-x}Sn_x)Se$ alloys, $\mu_h \sim 10^4$ cm^2/V sec and $D_h \sim 2 \times 10^2$ cm^2/sec. If $\tau_R = 10^{-11}$ sec, $W = 10$ μm, and $\mathcal{E}_0 = 3 \times 10^3$ V/cm, and if the power delivered by the laser is designated P_L, then the current is

$$i_h = A j_h(W) = 500 P_L \text{ mA} , \quad (35)$$

where P_L is measured in watts. [A is the area on which the light beam is focused; i_h is independent of A for the boundary condition (32b)]. Thus, even at modest cw laser power outputs, the photoinduced currents will be measurable. From conductivity measurements one can then determine τ_R (via l_h) as a function of the applied electric field. The role of the electric field is twofold: it induces carrier drift, and it changes τ_R via the effective gap.

Although we have concentrated on the experimental arrangement illustrated in Fig. 7, an experimentally better geometry is probably the one illustrated in Fig. 9, since the light beam falls on an area adjacent to the current probe and since the optical alignment can be adjusted to obtain the maximum photoinduced currents for a given laser output and applied field.

One might worry that in the suggested experiments the semiconductor would begin to lase due to the inverted electron-hole population and that this would interfere with the plasmon-assisted recombination. This need not be a problem. When the hole densities are in the range 10^{14}–10^{15} cm^{-3}, the semiconductor alloys we have been discussing lase only at quite low temperatures. The proposed experiments can be done at liquid-nitrogen or room temperature.

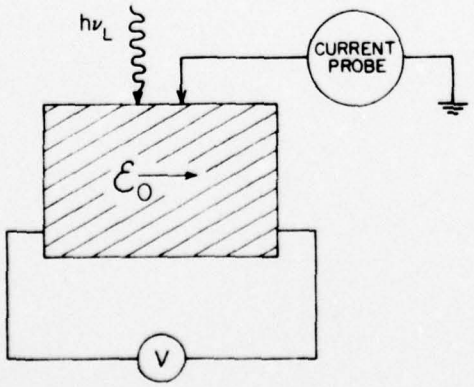

FIG. 9. Schematic of a photoconductivity experiment.

Plasmon-assisted electron-hole recombinations can be used in modulating the density-dependent parameters of the narrow-gap semiconductors. Nurmikko and Pratt suggested an application of this to modulate infrared radiation in picosecond range[27] which is of some practical interest. The light-absorption coefficient $\alpha(\nu)$ of a direct gap crystal is a relatively sensitive function of the carrier densities. If a large number of carriers are created by an intense light beam, $\alpha(\nu)$ decreases and the crystal becomes transparent. Transparency may increase by as much as three to five times, depending upon the pulse intensity. After the pulse, recombinations reduce the carrier density and eventually restore $\alpha(\nu)$ to its original value. For $(Pb_{1-x}Sn_x)Se$ and $(Pb_{1-x}Sn_x)Te$ crystals, the intense excitation pulses can have ν near the visible end of the spectrum. If $E_G \sim \hbar\omega_p$, these pulses produce picosecond transients in $\alpha(\nu)$ due to the fast recombinations. Therefore, if a light signal whose frequency is near the absorption edge ($h\nu \sim E_G$, which is far infrared) is sent through the sample as it is being pulsed by short-wavelength radiation, the transmitted infrared signal will be modulated in picosecond range. Nurmikko and Pratt appear to have observed these transients.

In summary, we have shown that in narrow-gap semiconductors the plasmon-assisted recombination is extremely rapid if the band gap and the plasma frequency are close. We have calculated the recombination rate and suggested experiments in which this type of recombination can be investigated. We have discussed its behavior in a constant electric field. We have also discussed a particular case in which the rapidity of the recombination might be usefully applied to devices.

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I wish to express my gratitude to P. A. Wolff for suggesting the problem. I thank D. Kobe for helpful comments on the presentation.

APPENDIX

In this Appendix, the integral involved in Eq. (17) is evaluated in the long-wavelength approximation at zero temperature. The purpose of the Appendix is essentially to illustrate the approximations involved in arriving at Eq. (19).

As plasmons decay, electronic transitions occur across the Fermi surface; therefore, for $q \to 0$, the energy denominators in Eq. (16) become

$$\frac{1}{\hbar\omega_q - E_c(\vec{k}+\vec{q}) + E_c(\vec{k})} - \frac{1}{\hbar\omega_q - E_c(\vec{k}+\vec{q}') + E_c(\vec{k}+\vec{q}'-\vec{q})} \simeq -\frac{1}{(\hbar\omega_p)^2}\,\vec{q}\cdot\left[\nabla_{\vec{k}}E_c(\vec{k}+\vec{q}') - \nabla_{\vec{k}}E_c(\vec{k})\right]$$

$$\simeq -\frac{1}{(\hbar\omega_p)^2}\,\vec{q}\cdot\vec{q}'\left(\frac{1}{k}\frac{dE_c(\vec{k})}{dk}\right)_{k=k_F} = \frac{-\vec{q}\cdot\vec{q}'}{m_{1s}\omega_p^2} \quad . \tag{A1}$$

Thus Eq. (17) for $\vec{q}=0$ becomes

$$\Delta(0) = \left(\frac{\hbar e^4 n_i}{6\pi^3\epsilon_\infty^2 m_{1s}^2 n_c \omega_p}\right)\int d\vec{k}\int d\vec{q}'\,\frac{(q')^2}{|(q')^2+\kappa^2|^2}\,f_{\vec{k}}^c(1-f_{\vec{k}+\vec{q}}^c)\delta(E_c(\vec{k}+\vec{q}')-E_c(\vec{k})-\hbar\omega_p)\,. \tag{A2}$$

Because of the δ function and (18), in the limit of zero temperature

$$f_{\vec{k}}^c(1-f_{\vec{k}+\vec{q}}^c)\delta(E_c(\vec{k}+\vec{q}')-E_c(\vec{k})-\hbar\omega_p) = \frac{1}{1+e^{\beta[E_c(\vec{k})-E_F]}}$$

$$\times\left(1-\frac{1}{1+e^{\beta[E_c(\vec{k})+\hbar\omega_p-E_F]}}\right)\delta(E_c(\vec{k}+\vec{q}')-E_c(\vec{k})-\hbar\omega_p)$$

$$\xrightarrow{T\to 0}\begin{cases}\delta(E_c(\vec{k}+\vec{q}')-E_c(\vec{k})-\hbar\omega_p) & \text{if } E_c(\vec{k})\le E_F\le E_c(\vec{k})+E_F,\\ 0 & \text{otherwise.}\end{cases} \tag{A3}$$

Thus, when conduction electrons are degenerate, Eq. (17) can be written

$$\Delta(0) \simeq \frac{\hbar e^4 n_i}{6\pi^3\epsilon_\infty^2 m_{1s}^2 n_c\omega_p}\int_{E_c(\vec{k})\le E_F\le E_c(\vec{k})+\hbar\omega_p} d\vec{k}\int d\vec{q}\,\frac{q^2\delta(E_c(\vec{k}+\vec{q})-E_c(\vec{k})-\hbar\omega_p)}{(q^2+\kappa^2)^2}\,, \tag{A4}$$

where we have dropped the prime on $\vec{q}$. When $E_c(\vec{k})$ in (4a) is substituted into (A4), the $\vec{q}$ integral becomes

$$2\pi\int_0^\infty dq\,q^2\int_{-1}^{+1} d(\cos\theta)\,\frac{q^2}{(q^2+\kappa^2)^2}$$

$$\times\,\delta\left(\left[\left(\frac{E_G}{2}\right)^2+\frac{\hbar^2E_G}{2m_{\rm BI}}(k^2+q^2+2kq\cos\theta)\right]^{1/2}-\left[\left(\frac{E_G}{2}\right)^2+\frac{\hbar^2E_Gk^2}{2m_{\rm BI}}\right]^{1/2}-\hbar\omega_p\right)$$

$$=\left(\frac{2\pi m_{\rm BI}}{\hbar^2E_Gk}\right)\left[2E_c(k)+2\hbar\omega_p-E_G\right]\int_{Q_-(k)}^{Q_+(k)} dq\,\frac{q^3}{(q^2+\kappa^2)^2}\,, \tag{A5}$$

where

$$Q_\pm(k) = \pm k + \left\{k^2+(2m_{\rm BI}\hbar\omega_p/\hbar E_G)\left[\hbar\omega_p - E_G + 2E_c(k)\right]\right\}^{1/2}\,. \tag{A6}$$

The remaining integral in (A5) is elementary

$$\int_{Q_-(k)}^{Q_+(k)} dq\,\frac{q^3}{(q^2+\kappa^2)^2} = \frac{1}{2}\left[\frac{Q_-^2}{\kappa^2+Q_-^2}-\frac{Q_+^2}{\kappa^2+Q_+^2}+\ln\left(\frac{\kappa^2+Q_+^2}{\kappa^2+Q_-^2}\right)\right]\,. \tag{A7}$$

To do the $\vec{k}$ integration, we note that the integrand does not depend on the direction of $\vec{k}$. Integration of the angular variables of $\vec{k}$ gives 4π. To evaluate the remaining integral, it is more convenient to change the integration variable from k to

$$E \equiv E_c(k) = \tfrac{1}{2}E_G + \left[(\tfrac{1}{2}E_G)^2+\hbar^2k^2E_G/2m_{\rm BI}\right]^{1/2}\,. \tag{A8}$$

In terms of E, Eq. (A6) becomes

$$Q_\pm(E) = (2m_{\rm BI}/\hbar^2E_G)^{1/2}\left\{\left[(E+\hbar\omega_p)(E+\hbar\omega_p-E_G)\right]^{1/2}\pm\left[E(E-E_G)\right]^{1/2}\right\}. \tag{A9}$$

Therefore Eq. (A4) becomes

$$\Delta(0)\simeq\left(\frac{2e^4n_im_{\rm BI}^2}{3\pi\epsilon_\infty^2m_{1s}n_c\omega_p\hbar^3E_G^2}\right)\int_{E_F-\hbar\omega_p}^{E_F} dE(2E-E_G)(2E+2\hbar\omega_p-E_G)\left[\frac{Q_-^2}{\kappa^2+Q_-^2}-\frac{Q_+^2}{\kappa^2+Q_+^2}+\ln\left(\frac{\kappa^2+Q_+^2}{\kappa^2+Q_-^2}\right)\right]. \tag{A10}$$

Note that E is confined to a thin shell at the Fermi surface: $E_F - \hbar\omega_p \leq E \leq E_F$. E is therefore large relative to E_G or $\hbar\omega_p$. Thus, to a good approximation, (A9) can be written

$$Q_+ \simeq (2m_{\rm BI}/\hbar^2 E_G)^{1/2}(2E + \hbar\omega_p - E_G)\,, \qquad \text{(A11)}$$

$$Q_- \simeq (2m_{\rm BI}/\hbar^2 E_G)^{1/2}(\hbar\omega_p)\,. \qquad \text{(A12)}$$

The logarithmic terms in (A10) can then be integrated by parts, and the other integrals are elementary. Integrating over E and substituting the value of $m_{\rm BI}/E_G$ and κ from (6) and (15), respectively, we obtain the expression given by (19).

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†Visiting.

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APPENDIX C

PICOSECOND OPTICAL ABSORPTION

AT 1.06 μm AND 1.55 μm IN THIN GERMANIUM SAMPLES AT

HIGH OPTICALLY-CREATED CARRIER DENSITIES

PICOSECOND OPTICAL ABSORPTION AT 1.06 μm AND 1.55 μm IN THIN GERMANIUM SAMPLES AT HIGH OPTICALLY-CREATED CARRIER DENSITIES

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Recently, the enhanced transmission of single, ultrashort optical pulses at 1.06 μm through germanium as a function of incident pulse energy has been measured [1]. In addition, the temporal evolution of this enhanced transmission has been determined on a picosecond time scale using the excite and probe technique [1,2]. The latter measurements reveal that the probe transmission increases for 50-100 psec following excitation with an intense optical pulse. ELCI et al. [3] have attributed this rise in the probe transmission to a cooling of the hot electron-hole plasma created by the excitation pulse. In sharp contrast to this interpretation, AUSTON et al. [4] have stated that they expect the energy relaxation time to be too short to account for the rise in the probe transmission. AUSTON et al. also suggest that enhanced free-hole [5] and Coulomb-assisted indirect absorption [6] effects can be significant at the high optically-created carrier densities encountered in the excitation-probe measurements at 1.06 μm. Indeed, they suggest that these processes could introduce a minimum in the absorption versus carrier density relationship. AUSTON and MCAFEE [7] have proposed an alternative explanation for the temporal evolution of the probe transmission by combining the details of the way the absorption saturates as a function of carrier density with a monotonic decrease in carrier density with time due to Auger recombination [8]. The rise in probe transmission with time can then be explained in the following manner. We denote the density at which the minimum absorption occurs as n_{min}. The absorption of the excitation pulse creates an initial carrier density greater than n_{min}. As the carrier density is decreased by Auger recombination, the absorption coefficient will decrease in time until the carrier density reaches n_{min}, then increase. Thus, the probe transmission will increase then decrease if the initial, optically-created carrier density is greater than n_{min}. We stress that the success of the second model as it now stands depends on the absorption decreasing then increasing with carrier density: there must be an absorption minimum.

Here, we report measurements of the combined free-carrier, intervalence band free-hole, and indirect absorbance in thin germanium samples at a wavelength of 1.55 μm during excite and probe experiments at a wavelength of 1.06 μm. Our interests in these measurements are twofold. First, we want to ascertain whether or not free-carrier, free-hole, and indirect absorption effects are important in excite-probe experiments at 1.06 μm. Second, if these effects are important, can they, together with Auger effects, account for the rise in probe transmission.

The experimental configuration is depicted in Fig.1. This arrangement is similar to the arrangement utilized by AUSTON et al. [8]. In this application of the excite and probe technique, a high density plasma is created by direct

absorption of an intense excitation pulse, and the evolution of the plasma is monitored by a second probe pulse. The excitation pulses were selected by a laser-triggered spark gap and a Pockel's cell from trains of pulses produced by a mode-locked Nd:glass laser. The pulses were 5 to 10 psec in duration and had peak powers of approximately 10^8 watts at a wavelength of 1.06 μm, and they produced a measured irradiance of approximately 4×10^{-3} J/cm^2 when focused on the crystal surface. The plasma produced by the absorption of the excitation pulse was probed using weak pulses of two types: one had an energy greater than the direct energy band gap for germanium and the other had an energy less than the direct gap but greater than the indirect gap. The former was derived from the excitation pulse using a beam splitter as shown in Fig.1. The latter,

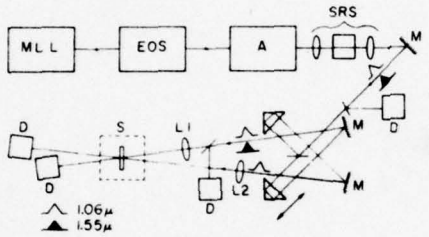

Fig.1 Block diagram of the experimental configuration for excite and probe measurements at 1.06 μm and 1.55 μm, where MLL denotes the mode-locked laser, EOS the electro-optic switch, A the laser amplifier, SRS the Stimulated-Raman-Scattering cell, M a mirror, D a detector, L1 and L2 lens, and S the sample

having a wavelength of 1.55 μm, was produced by stimulated Raman scattering in benzene. We emphasize that the energy of a quanta at 1.06 μm (1.17 ev) is sufficient to excite direct band-to-band transitions in germanium as well as free-carrier, free-hole, and indirect transitions; whereas, the energy of a quanta at 1.55 μm (0.80 ev) falls below the direct band gap but above the indirect gap and is, thus, only a measure of the combined free-carrier, free-hole, and indirect processes. The incident excitation pulse irradiance was measured, and the overlap of excitation and probe pulses was ensured, employing techniques described in [8].

The germanium sample was a high purity ($\rho_{min} = 40\,\Omega$ cm) single crystal cut with the (111) plane as face. The sample was polished and etched with Syton to a thickness of 6 μm as determined by interferometric techniques.

The results of the measurement of the change in absorbance of the thin germanium crystal as a function of increasing carrier number (incident excitation pulse energy at 1.06 μm) are shown in Fig.2 for photon energies of 1.17 and

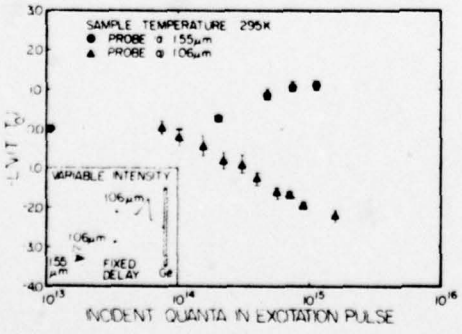

Fig.2 Change in absorbance, $-\ell n(T/T_0)$, of the germanium sample at 1.06 μm and 1.55 μm as a function of incident excitation pulse energy at 1.06 μm, where T_0 is the linear transmission of the sample at the wavelength under consideration

0.8 ev. The data were obtained in the following manner. The crystal was illuminated by variable energy optical pulses of wavelength 1.06 μm, and the transmission of each pulse was measured. Each pulse at 1.06 μm was followed immediately (at a fixed delay of 15 psec) by pulses that monitored the absorbance of the crystal at wavelengths of 1.55 μm and 1.06 μm. The optical absorbance at 1.17 ev is seen to decrease by approximately 2.3 as the carrier number increases. This corresponds to a transmission increase of an order of magnitude. By contrast, the absorbance at 0.8 ev increases roughly by 1.0, corresponding to a decrease in transmission of a factor of 2.7. Each data point shown is the average of at least ten separate observations. The data were very reproducible within the error bars.

A striking feature of the data presented in Fig.2 is that the absorbance of the crystal at 1.06 μm does not decrease then increase as required by the model suggested by AUSTON and MCAFEE [7]. In fact, as can be seen from Fig.2, any decrease in carrier density with time caused by carrier recombination will be accompanied by an increase in the total absorbance at 1.06 μm. Thus, a temporal decay of carrier density alone can not be combined with the absorption versus density relationship to account for the rise in probe transmission at 1.06 μm. In addition, the change in absorbance at 0.8 ev, which is sensitive to free-carrier, free-hole, and indirect absorption effects, is smaller in magnitude and opposite in sign to that measured at 1.17 ev, which is sensitive to direct absorption effects as well. Although care must be taken when extrapolating absorbance measurements at 0.8 ev to 1.17 ev, we believe that the sign and magnitude of this absorbance change at 0.8 ev and the observed monotonic decrease in overall absorbance at 1.17 ev suggests that the saturation of the absorbance at 1.17 ev is dominated by changes in the direct absorption coefficient.

The results of excite-probe experiments that measure the temporal evolution of these changes in absorbance are presented in Fig.3. In this experiment the sample was irradiated by an optical pulse at 1.06 μm containing roughly 10^{15} quanta and was probed at various delays by weak pulses having wavelengths of 1.55 μm and 1.06 μm. The transmission of the probe pulse at 1.06 μm initially increases as the delay between the two pulses increases, reaching a peak value at a delay of approximately 50 psec. Meanwhile, the absorbance of the sample

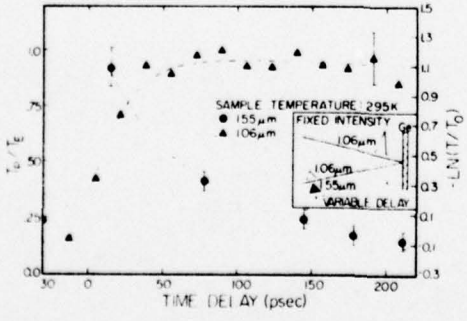

Fig.3 Probe pulse absorption versus delay between the excitation pulse at 1.06 μm and 1.55 μm. The probe data at 1.06 μm is plotted as the normalized ratio of probe pulse transmission to excitation pulse transmission, T_p/T_E, and the data at 1.55 μm as the change in absorbance, $-\ell n(T_p/T_0)$, where T_0 is the linear transmission of the probe pulse at this wavelength

at 1.55 μm decreases by roughly 1.0 corresponding to a transmission increase of approximately 2.7. Different units were chosen for the ordinates of the 1.06 μm and 1.55 μm curves to facilitate presentation of both curves in the same figure. The probe measurements at 1.06 μm are identical to those performed by SMIRL et al. [1]. Also, the results of the probe measurements at

1.55 μm are similar to those obtained by AUSTON et al. [8]. However, these authors stated that they performed their measurements at excitation intensities such that the absorption of the excitation pulse was linear. Our experiments are clearly performed in the nonlinear region. In addition, the measurements of AUSTON et al. were performed on a 300 μm-thick sample; our sample was 6 μm-thick. Thus, our experimental configuration is almost identical to that employed in our original excite and probe experiments at 1.06 μm.

The measurements presented in Fig.3 indicate that free-carrier, free-hole, and indirect absorption can be significant at the carrier densities encountered during the excite and probe experiments at 1.06 μm presented here. Note that the increase with time of the probe transmission at 0.8 ev caused by the decrease in free-carrier, free-hole, and indirect absorbance is of the proper magnitude (if extrapolated unchanged to 1.17 ev) to account for a major portion of the probe rise at 1.17 ev. AUSTON et al. [8] attribute this decrease of the probe pulse absorbance at 1.55 μm with delay to a decrease in free-carrier absorption caused by a temporal decay in carrier density due to Auger recombination. The present experiments only allow the measurement of the change in the combined free-carrier, free-hole, and indirect absorbance, and they do not provide for a convenient separation of their individual contributions. In addition, we feel that the decrease in absorbance at 1.55 μm with delay could be accounted for by a number of mechanisms. One of these possible mechanisms is carrier reduction by Auger recombination. Another possibility, however, is that the decrease in absorbance at 1.55 μm is caused by a cooling of a hot carrier distribution created by the excitation pulse at 1.06 μm. This decrease in absorbance could be attributed to a decrease in free-carrier absorption and a decrease in the available states for indirect absorption as the large, hot carrier distribution cools.

In summary, the measurement of the transmission of thin germanium samples at 1.55 μm and 1.06 μm as a function of optically-created carrier densities indicates, over the range of densities encountered in these experiments, that the absorption versus density relationship at 1.17 ev does not exhibit a minimum and suggests that the combined free-carrier, free-hole, and indirect absorbance changes are opposite in sign and smaller in magnitude than the changes caused by saturation of the direct absorption. Second, the excite and probe measurements at these two wavelengths indicate that free-carrier, free-hole, and indirect absorption processes could be significant in interpreting excite and probe measurements at 1.06 μm.

The authors wish to acknowledge their debt to Dave Auston and Sigrid McAfee of Bell Laboratories for suggesting and sharing the model presented in this paper and for their helpful discussions.

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APPENDIX D

PICOSECOND OPTICAL MEASUREMENT OF FREE-CARRIER, INTERVALENCE-BAND, AND INDIRECT ABSORPTION IN GERMANIUM AT HIGH OPTICALLY-CREATED CARRIER DENSITIES

Picosecond optical measurement of free-carrier, intervalence-band, and indirect absorption in germanium at high optically-created carrier densities

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ABSTRACT

It has been suggested that enhanced intervalence-band and Coulomb-assisted indirect absorption effects may be significant at the high optically-created carrier densities encountered in the recent excite and probe experiments performed in germanium using intense picosecond optical pulses with a wavelength of 1.06 μm and that these processes may result in an absorption versus carrier density curve containing a minimum. Such a curve could then be combined with a recombination process to explain the results of the excite and probe experiments. Here, we report measurement of the combined free-carrier, intervalence-band, and indirect absorbance in thin germanium samples during these excite and probe experiments by exciting at 1.06 μm and by probing both at 1.06 μm and with a Raman-generated probe at 1.55 μm. The measurements suggest that these processes are significant at the high optically-created carrier densities encountered in the present excite and probe experiments; however, they do not introduce a minimum in the absorption versus carrier density curve as originally suggested.

I. INTRODUCTION

Recently, studies of the optical properties of high-density electron-hole plasmas generated in undoped germanium by intense, ultrashort pulses have provided direct information concerning ultrafast electronic processes.[1-12] Among these studies are the measurement of the enhanced transmission of single ultrashort optical pulses through germanium,[1,5] and the measurement of the temporal evolution of this enhanced transmission on a picosecond time scale using the excite and probe technique.[3,5] In the first of these experiments, the nonlinear transmission of a single picosecond 1.06 μm pulse was investigated as a function of incident optical pulse energy for sample temperatures of 100 K and 297 K. The resulting data, reproduced from Smirl et al.,[5] are shown in Fig. 1 along with theoretical curves to be discussed later. In the second study, the sample was first irradiated by an excitation pulse of sufficient energy to cause the transmission of the germanium to be enhanced. This initial pulse was then followed at various time delays by a weak probe pulse, of the same wavelength, that measured the temporal evolution of the germanium sample. Probe pulse transmission data for various delay times and for sample temperatures of 100 K and 297 K are presented in Fig. 2, again taken from Smirl et al.[5] The latter measurements reveal that the probe transmission increases for approximately 100 psec following excitation for a sample temperature of 100 K; however, the rise in probe transmission is less than 40 psec at 297 K. Originally, Shank and Auston[3] attributed this rise in the probe transmission to a saturation of the absorption

caused by a filling of the optically-coupled conduction-band states and a depletion of the valence-band states by direct band-to-band transitions induced by the excitation pulse. Thus, the optically-created carrier density, and consequently the increase in probe transmission, follows the integrated optical pulse energy. This interpretation was based on observations performed only at room temperature. As we shall later demonstrate, the rise in probe transmission at room temperature is indeed indistinguishable from other integration effects, in agreement with this interpretation. However, such a model cannot account for the slower rise observed at 100 K. Elci et al.,[7] in a recently proposed model, have attributed this rise in the probe transmission to a cooling of a hot electron-hole plasma created by the excitation pulse. In sharp contrast to this interpretation, Auston et al.[12] have stated that they expect the energy relaxation time to be too short to account for the rise in probe transmission. Indeed, Auston and McAfee[13] have suggested a plausible alternative explanation for the temporal evolution of the probe transmission in terms of enhanced Coulomb-assisted indirect absorption, intervalence-band absorption, and Auger recombination. This explanation does not require hot electron effects.

Here, we report measurements of the combined free-carrier, intervalence-band, and indirect absorbance in thin germanium samples at a wavelength of 1.55 μm during excite and probe experiments at a wavelength of 1.06 μm. Our interests in these measurements are twofold. First, we want to ascertain whether or not free-carrier, intervalence-band, and indirect absorption effects are important in excite-probe experiments at 1.06 μm. Second, if these effects are important, can they, together with Auger effects, account for the rise in probe transmission.

In the next section, we briefly review the model presented by Elci et al.[7] and emphasize the processes, or omission of processes, that are relevant to the present measurements. In addition, in Sec. III, we briefly describe a model suggested by Auston and McAfee[13] that accounts for the rise in probe transmission without employing hot electron effects. These reviews are then followed in Sec. IV by a description of the experimental apparatus and techniques for measuring the combined contributions of free-carrier, intervalence-band, and indirect absorption processes to the excite and probe response of germanium. Section V presents a discussion of our results, and the final section, Sec. VI, our conclusions.

II. COMMENTS ON THE HOT ELECTRON MODEL

Briefly, according to the model proposed by Elci et al., the transmission of a single optical pulse through a thin (5.2-μm-thick) germanium sample as a function of incident pulse energy (Fig. 1) and the transmission of a weak probe pulse as a function of time delay after an energetic pulse (Fig. 2) can be accounted for in terms of direct band-to-band absorption, free-carrier absorption, phonon-assisted intervalley scattering, phonon-assisted carrier relaxation, carrier-carrier collisions and nonradiative recombination in the following manner. When an excitation pulse is incident on the germanium sample, the unreflected portion of the pulse enters the sample where most of it is absorbed by direct transitions, creating a large density of electrons (holes) in the central valley of the conduction (valence) band. The electrons are rapidly ($<10^{-14}$ sec) scattered to the conduction band side valleys by long wave vector phonons. Carrier-carrier scattering

events, which occur at a rate comparable to the direct absorption rate, ensure that the carrier distributions are Fermi-like and that both electron and hole distributions have the same temperature, which can be different from the lattice temperature. Since the photon energy $\hbar\omega_0$ is greater than either the direct energy gap E_0 or the indirect band gap E_G, such a direct absorption event followed by phonon-assisted scattering of an electron to the side valleys results in the photon giving an excess energy of $\hbar\omega_0 - E_G$ to thermal agitation. This excess energy results in an initial distribution temperature (approximately 1500 K for a lattice temperature of 300 K) due to direct absorption that is greater than the lattice temperature. Thus, the single pulse transmission would begin at its Beer's law value and increase as a function of incident optical pulse energy because of the partial filling (depletion) of the optically-coupled states in the conduction (valence) band as a result of direct absorption. Other processes such as free-carrier absorption and nonradiative recombination events (i.e., Auger and plasmon-assisted recombination) can further raise the carrier temperature during the passage of the excitation pulse, while phonon-assisted intravalley relaxation processes can reduce the carrier temperature.

After the passage of the excitation pulse, the interaction region of the sample contains a large number of carriers (10^{19}-10^{20} cm^{-3}) with a high distribution temperature. The final temperature is determined by the number of quanta in the excitation pulse and the relative strengths of the nonradiative recombination and the phonon-assisted relaxation rates as discussed by Latham et al.[10] As time progresses, the distribution will continue to cool by phonon-assisted intravalley relaxation. Experimentally, the probe pulse interrogates the evolution of the

distribution after the passage of the excitation pulse and is a sensitive measure of whether the optically-coupled states are available for absorption or are occupied. Immediately after the passage of the excitation pulse, the probe transmission is small since the electrons (holes) are located high (low) in the conduction (valence) bands because of the high distribution temperature, leaving the states that are optically coupled available for direct absorption. Later, as the distribution temperature cools and carriers fill the states needed for absorption, the transmission increases. The subsequent slow fall in probe transmission at longer delays, as seen in Fig. 2, is attributed to carrier recombination, which reduces the carrier density and once again frees the optically-coupled states for absorption, and to diffusion.[3,11]

The theoretical fits from Elci et al.[7] to the single pulse transmission data and probe pulse data are shown as dashed lines in Fig. 1 and Fig. 2. Given the complexity of the problem, the overall fit can be regarded as satisfactory. Nonlinear transmission measurements in which the energy band gap of the germanium sample was tuned by hydrostatic pressure[8] have been accounted for by this model as well.

Despite the apparent successes of this model, some basic questions remain concerning the roles of the various physical processes in determining the saturation and temporal evolution of the optical transmission of thin germanium samples under intense optical excitations. Elci et al.[7] noted that their calculations included only a limited number of the possible electronic interactions and contained serious assumptions that warranted further theoretical and experimental investigation. The major assumptions were the following: (1) The carrier-carrier collision rate was assumed to be high enough to justify taking the carrier distributions

to be Fermi-Dirac. Ferry[14] has recently reexamined this approximation by calculating the time and energy dependence of the distribution function at the high carrier photogeneration rates encountered here. He concludes that on a time scale of tens of picoseconds the distribution function does indeed approximate a Fermi distribution; however, on shorter time scales it contains a δ-function-like spike located at the optically-coupled states. Thus, for purposes of calculating the probe pulse transmission, one may reasonably assume the distribution is Fermi-like. (2) Carrier Fermi energies and temperatures were taken to depend only on time, rather than on both space and time, thus ignoring the pulse propagation and carrier diffusion problems within the optical interaction region of the sample. Therefore, parameters describing the electron-hole plasma, such as the electron number, must be viewed as spatial averages throughout the sample volume. Elci et al.[11] have recently extended their previous model, through a simple calculation, to indicate the possible effects of carrier diffusion on these optical measurements.

Elci et al.[7] noted at the outset that their work contained only a few of the many possible electronic interactions. Recent studies[4,12-14] indicate that processes other than those named above may be important. Most of these effects, such as band-gap narrowing,[14] intervalence-band absorption,[13] Auger recombination[4] and Coulomb-assisted indirect absorption,[12] are only observed at large carrier densities. The possible importance of including these processes in any interpretation of the rise in probe transmission is demonstrated in the following sections. One of the main objectives of the present study is to investigate the effects of the combined intervalence-band, Coulomb-assisted indirect,

and free-carrier absorption and Auger recombination on the excite and probe measurements at 1.06 μm.

In the previous two paragraphs, we have outlined the assumptions and omissions of the initial hot electron model; however, there is another problem associated with the original calculations that is of importance to the present work. The physical constants for germanium, specifically the electron-phonon coupling constants, are not well enough known to allow a precise calculation of the energy relaxation rate. Latham et al.[10] have previously discussed this point in detail. For the theoretical fits shown in Fig. 2, the electron-phonon coupling constants are chosen as 6×10 erg-cm^{-1} for a lattice temperature of 297 K and 2×10^{-4} erg-cm^{-1} at 100 K. These values are within the range of the accepted theoretically and experimentally determined values listed by Latham et al.;[10] however, they are much lower than the mean value of 1×10^{-3} erg-cm^{-1} as obtained from an average of the eight values listed. Since the carrier cooling rate is proportional to the square of the electron-phonon coupling constant, the fitted values result in carrier cooling rates that are 3 and 25 times slower than that obtained by using the average value. In fact, a repetition of the original calculations substituting the average electron-phonon coupling constant shows (see Latham et al., Fig. 8) that carrier cooling is too rapid to account for the rise in probe transmission, in complete agreement with the claims by Auston et al.[12] However, van Driel[15] has recently calculated the influence of hot phonons on the carrier energy relaxation rate in these problems. These calculations suggest that the long equilibration time for the hot carriers is due to a relaxation bottleneck produced by a buildup of the optical phonon population on a picosecond time scale.

The results of these calculations, taking into account optical phonon heating and employing only a single average temperature-independent electron-phonon coupling constant, are shown as solid curves in Fig. 2. Notice that the inclusion of hot phonons accounts for one of the major discrepancies between the original theory and experiment. Namely, in contrast to the original theory that predicted a delayed, steep rise, the present theory shows a steep rise with gradual leveling off in agreement with the data. The solid curves in Fig. 2 were taken directly from van Driel.[15] The agreement between the modified theory and experiment is remarkable; however, this agreement should be regarded as somewhat fortuitous in view of the simplifications of the model, the limited number of processes included, and the uncertainty in many of the physical constants.

For emphasis, we now summarize the principal points of this section: (1) According to Elci et al., the rise of the optical transmission of the germanium with time as monitored by a weak probe pulse is attributed to the cooling of a hot electron-hole plasma, created by the absorption of the excitation pulse. (2) In view of the above discussions and in the absence of any direct experimental evidence, the authors regard the question of the magnitude of the energy relaxation rate and the question of the origin of the rise of the probe transmission as open. It is in this spirit of suggesting plausible alternative models for evaluation that McAfee and Auston[13] first suggested the possibility of describing the rise in probe transmission in terms of free-carrier, intervalence-band, and Coulomb-assisted indirect absorption together with Auger recombination. This model is reviewed in the next section.

III. INDIRECT, INTERVALENCE-BAND, AND FREE-CARRIER ABSORPTION AND AUGER RECOMBINATION

In direct contrast to the above model, Auston and McAfee[13] have recently suggested an alternative explanation for the delayed probe pulse transmission in germanium without requiring the introduction of hot electron effects. In fact, their suggested explanation is based on three of the processes neglected by the original calculations of Elci et al.:[7] Auger recombination, Coulomb-assisted indirect absorption, and intervalence-band absorption. Specifically, this model is based on experiments by Auston et al.,[4] on 300-μm-thick samples, that are interpreted as demonstrating that Auger recombination effects are important on picosecond time scales; on the observation of enhanced indirect Coulomb-assisted absorption in heavily-doped n-type germanium by Haas;[16] and on the observation of strong intervalence-band absorption between the light and heavy-hole and split-off valence bands in p-type germanium by Newman and Tyler.[17] Since the present work is an attempt to experimentally investigate the role of these processes in determining the evolution of the optical properties of germanium at high carrier densities, we shall briefly state the conclusions of these two works. We then review the manner in which enhanced indirect absorption and intervalence-band absorption together with Auger recombination could account for the probe transmission versus time delay after an intense excitation pulse in thin germanium samples.

Haas[16] reports that the indirect absorption rises more rapidly with photon energy in n-type germanium than in pure germanium. The

effects considered as sources for this extra absorption are: (1) modification of the band structure by impurities, (2) impurity-assisted indirect transitions and (3) Coulomb-assisted indirect transitions, where the virtual scattering of the electrons from the central to the side valley is by electron-electron scattering. Haas concludes that at high concentrations electron-electron scattering dominates the indirect transitions. If we extrapolate his results, they suggest that, at an optical wavelength of 1.06 μm and electron densities as large as 2×10^{20} cm^{-3}, the indirect absorption coefficient might be as large as 10^4 cm^{-1}.

Newman and Tyler[17] report measurement of the free-hole absorption in p-type germanium as a function of impurity and carrier concentration. The effects to be considered in explaining the strong observed free-hole absorption at high carrier concentrations are: (1) modification of the band structure by impurities, (2) band-to-band transitions between light and heavy-hole valence bands and the split-off valence band as the position of the Fermi level changes within the valence band with doping, and (3) indirect transitions induced by charged impurity centers. They conclude that their observations suggest that both intervalence band transitions and impurity-assisted indirect transitions contribute. In experiments employing undoped samples, where the carriers are generated by optical absorption, only the intervalence-band process will be significant. If their results are extrapolated to a photon energy of 1.17 ev and a concentration of 2×10^{20} cm^{-3}, the free-hole absorption coefficient could again be as large as 10^4 cm^{-1}.

The suggestion of an alternate explanation for the delayed probe transmission of optically-excited germanium without requiring phonon-assisted relaxation of hot electrons is based on the two mechanisms

discussed above. In particular, it is based on the details of the way germanium absorption might vary with increasing optically-created carrier density. Consider the behavior of the total absorption coefficient as the carrier density increases because of band-to-band transitions during the passage of an intense excitation pulse. (The carrier temperature is taken to be that of the lattice, since all carrier energy relaxation processes are assumed to be too rapid for observation, in contrast to Elci et al.[7]) The direct absorption coefficient will decrease with increasing density as optically-created electrons (holes) clog the states needed for optical absorption in the conduction (valence) band. Meanwhile, as the density increases, the additional mechanisms discussed previously, Coulomb-assisted indirect absorption, intervalence-band absorption and free-carrier absorption, increase. Thus, the absorption coefficient could initially decrease with increasing density, as the direct absorption coefficient saturates, then increase with increasing density, as the free-carrier, intervalence-band, and indirect absorption coeffients dominate. In short, the fact that the direct absorption coefficient decreases with increasing carrier number and that the enhanced intervalence-band, free-carrier, and Coulomb-assisted indirect absorption coefficients increase can result in introducing a minimum in the absorption versus density relationship as suggested by Auston et al.[12] We denote the density at which the minimum absorption might occur as n_{min}. The rise in the probe transmission with time can now be accounted for by combining the details of the way the absorption saturates with carrier density together with a monotonic decrease in carrier density with time due to Auger recombination in the following manner. The absorption of the excitation pulse creates an initial carrier

density greater than n_{min}. As the carrier density is decreased by Auger recombination, the absorption coefficient of the sample will decrease in time until the carrier density reaches n_{min}, then increase. Thus, the probe transmission will increase then decrease if the initial, optically-created carrier density is greater than n_{min}. We shall henceforth refer to the model described in this section as the recombination model because of the role of the Auger recombination.

For emphasis, we summarize the features of the two models that are essential for comparison with present measurements. We note, once again, that Elci et al.[7] attribute the rise in probe transmission with delay after an excitation pulse to phonon-assisted cooling of a hot carrier distribution and that free-carrier, intervalence-band, and Coulomb-assisted indirect transitions were either omitted from the model or judged to be insignificant. Auger recombination processes were omitted from this model. The recombination model accounts for the rise in probe transmission by combining Auger recombination with an absorption versus density relationship containing a minimum. We stress that the success of the second model, as it now stands, depends on the absorption decreasing then increasing with carrier density: there must be an absorption minimum.

IV. EXPERIMENT

The particular experimental configuration used to measure the contributions of intervalence-band, free-carrier, and indirect absorption to the generation and evolution of dense, optically-created electron-hole plasmas in thin germanium samples is depicted in Fig. 3. This arrangement is similar to the arrangement utilized by Auston et al.[4]

In this application of the excite and probe technique, a high density plasma is created by direct absorption of an intense excitation pulse, and the evolution of the plasma is monitored by a second probe pulse. The excitation pulses were selected by a laser-triggered spark gap and a Pockel's cell from trains of pulses produced by a mode-locked Nd:glass laser. The pulses were 5 to 10 psec in duration and had peak powers of approximately 10^8 watts at a wavelength of 1.06 μm, and they produced a measured irradiance of approximately 1×10^{-2} J/cm^2 when focused on the crystal surface. The plasma produced by the absorption of the excitation pulse was probed using weak pulses of two types: one had an energy greater than the direct energy band gap for germanium, and the other had an energy less than the direct gap but greater than the indirect gap. The former was derived from the excitation pulse using a beam splitter as shown in Fig. 3. The latter, having a wavelength of 1.55 μm, was produced by stimulated Raman scattering in benzene. The desired probe wavelength was selected by employing either a thick wafer of silicon to reject the 1.06 μm radiation or narrow band-pass optical filters to reject the 1.55 μm radiation. Other wavelengths generated by the stimulated scattering in benzene, such as 1.18 μm, were rejected by carefully selected interference filters. We emphasize that the energy of a quanta at 1.06 μm (1.17 ev) is sufficient to excite direct band-to-band transitions in germanium as well as free-carrier, intervalence-band, and indirect transition; whereas, the energy of a quanta at 1.55 μm (0.80 ev) falls below the direct band gap but above the indirect gap and is, thus, only a measure of the combined free-carrier, intervalence-band, and indirect processes. The incident excitation pulse irradiance was measured and the overlap of excitation

and probe pulses was ensured employing techniques described in Ref. 4. The excitation pulse irradiance was determined by measuring the energy transmitted through a pinhole located at the focus of the excitation beam and coplanar with the germanium wafer using a calibrated detector. The probe beam was more tightly focused than the excitation beam to ensure complete spatial overlap with the excitation beam. The size of the pinhole was such that it transmitted 50% of the excitation pulse and 90% of the probe. Despite these precautions, we observed indications of day-to-day variations in excitation-beam and probe-beam overlap. We attribute these variations to "hot" spots in the focused multimode laser pulses.

The germanium sample was a high purity ($\rho_{min} \approx 40$ Ω-cm) single crystal cut with the (111) plane as face. The sample was polished and etched with Syton to a thickness of 6 μm as determined by interferometric techniques.

V. RESULTS AND DISCUSSION

Here, we present and discuss the result of three separate measurements. (1) In the first of these, the sample is illuminated by an intense excitation pulse at a wavelength of 1.06 μm. The transmission of a weak probe pulse at 1.06 μm that arrives at a variable, delayed time after the excitation pulse is then monitored. We perform these measurements, which repeat those by Smirl et al.,[5] to ensure that the rise in probe transmission can be separated from any artifacts of the measurement technique. (2) Next, the sample is irradiated by 1.06 μm-excitation pulses of various intensity that create electron-hole plasmas of varying density by direct band-to-band transitions. The change in

absorbance at 0.80 ev and 1.17 ev, as a function of plasma density, is then measured by monitoring the transmission of weak probe pulses at 1.55 μm and 1.06 μm that arrive a short fixed delay after excitation. These 1.06-μm-probe transmission measurements will provide the absorbance versus density curve needed for investigation of the recombination model described in Section III. The 1.55-μm-probe measurements will give a measure of the importance of free-carrier, intervalence-band, and indirect absorbance as a function of carrier density. (3) Finally, the transmission of a weak 1.55-μm- probe pulse is measured at various delays following an intense 1.06-μm-excitation pulse. The 1.55-μm-probe pulse monitors the temporal evolution of the change in the combined free-carrier, intervalence-band, and indirect absorbance following the photogeneration of a dense electron-hole plasma.

The results of experiments that measure the temporal evolution of the transmission of a thin germanium sample at a wavelength of 1.06 μm following the creation of a dense electron-hole plasma are shown in Fig. 4. The measurements were performed in the following manner (see insert, Fig. 4). The sample was irradiated by 1.06-μm-excitation pulses containing approximately 2×10^{15} quanta, and the transmission of each pulse was measured. Each excitation pulse was then followed at various delays by a weak probe pulse at 1.06 μm. These measurements were performed for sample temperatures of 100 K and 295 K. The data are plotted as the ratio of probe pulse transmission T_P to excitation pulse transmission T_E, in arbitrary units, versus time delay in picoseconds. The arbitrary units are chosen so that the peak of the probe transmission at 100 K is unity. The actual value of the ratio T_P/T_E was observed to be as large as six; however, this value strongly depends on the quality of

the spatial overlap of the focused excitation and probe pulses on the sample surface. These measurements are identical to those performed by Smirl et al., as presented in Fig. 2. However, when comparing the two sets of data, one must realize that the sample thicknesses and focused optical spot sizes are not identical.

The measurements of Smirl et al.[5] are repeated so that we can more carefully investigate the possibility that the rise in probe transmission follows the integrated optical energy. Specifically, we want to be assured that the rise in probe transmission is a real effect and that it is not an artifact of the excite-probe technique that can be attributed to the finite width of the optical pulses. For comparison, we have calculated the probe pulse transmission by assuming a Drude model for the electron-hole plasma, calculating the optical polarization, and substituting into the wave equation. The details of such a procedure are published elsewhere,[3,18] and they are not repeated here. The calculated rise in the probe pulse transmission, neglecting all decay processes, is simply proportional to the integral of the pulse autocorrelation function. The resulting theoretical integration curve assuming Gaussian-shaped optical pulses of 10 psec width (FWHM) is shown as a solid line in Fig. 5. Experimental data from Fig. 4 are plotted on an expanded time scale for comparison. The authors conclude that the experimental rise in probe transmission at 295 K is indistinguishable from integration effects, in agreement with the original interpretation of the room temperature data by Shank and Auston.[3] However, the rise at 100 K cannot be attributed to such effects and represents a physical effect. It is this rise in probe transmission at 100 K that is the object of our investigation. Finally, we note that coherent coupling effects[3,18] are

observed in these experiments as well; however, the delay increments of Fig. 5 are too coarse to resolve them.

The results of the measurement of the change in absorbance of the thin germanium crystal as a function of increasing carrier number (incident excitation pulse energy at 1.06 μm) are shown in Fig. 6 for photon energies of 1.17 and 0.8 ev. These data were obtained in the following manner. The crystal was illuminated by variable energy pulses with a wavelength of 1.06 μm, and the transmission of each pulse was measured. Each pulse at 1.06 μm was followed immediately (at fixed delays of 17 and 26 psec) by pulses that monitored the absorbance of the crystal at wavelengths of 1.55 μm and 1.06 μm. The optical absorbance at 1.17 ev is seen to decrease by approximately 3.5 as the carrier number increases. This corresponds to a transmission increase by a factor of 30. By contrast, the absorbance at 0.8 ev increases roughly by 2.3, corresponding to a decrease in transmission by approximately an order of magnitude. Each data point shown is the average of at least eight separate observations. The data were very reproducible within the error bars.

A striking feature of the data presented in Fig. 6 is that the absorbance of the crystal at 1.06 μm does not decrease then increase as required by the recombination model of Sec. III. In fact, as can be seen from Fig. 6, any decrease in carrier density with time caused by carrier recombination will be accompanied by an increase in the total absorbance at 1.06 μm. Thus, a temporal decay of carrier density alone cannot be combined with the absorption versus density relationship to account for the rise in probe transmission at 1.06 μm. The total change in absorbance of the crystal at 1.06 μm as the carrier density is

increased is given by

$$-\ell n(T/T_0) = \int_0^{\ell} \Delta\alpha_{DA}(x)dx + \int_0^{\ell} [\Delta\alpha_{FC}(x) + \Delta\alpha_{IB}(x) + \Delta\alpha_{ID}(x)]dx \quad . \quad (1)$$

where T_0 is the linear transmission of the sample; $\Delta\alpha_{DA}(x)$ is the change in the direct absorption coefficient caused by the increased carrier number at the position x into the crystal; $\Delta\alpha_{FC}(x)$, $\Delta\alpha_{IB}(x)$, and $\Delta\alpha_{ID}(x)$ are the changes in the absorption coefficient caused by free-carrier, intervalence-band, and indirect absorption, respectively; and ℓ is the crystal thickness. The electron density and, consequently, the absorption coefficients are allowed to depend upon position. The last term on the right hand of Eq. (1) will be positive since free-carrier, intervalence-band, and Coulomb-assisted indirect absorption coefficients all increase with carrier density, and the first will be negative because of the partial saturation of the available optically-coupled electronic states as the density increases. Since the overall absorbance at 1.17 ev is observed to monotonically decrease, we conclude that the saturation of the absorption is dominated by changes in the direct absorption coefficient. Although the free-carrier, intervalence-band, and indirect absorbance changes are smaller in magnitude and opposite in sign to those caused by saturation of the direct absorption, it is possible for them to significantly affect the overall magnitude of the total absorbance change. Inspection of Eq. (1) reveals that omission of these processes would result in a more rapid decrease in total absorbance with increasing carrier number than when they are included.

The measurement of the change in absorbance at 0.8 ev as the carrier number is increased by direct absorption of excitation pulses at 1.17 ev, as displayed in Fig. 6, tends to substantiate the arguments

of the previous paragraph. That is, the change in absorbance at 0.8 ev, which is sensitive to free-carrier, intervalence-band, and indirect absorption effects, is slightly smaller in magnitude and opposite in sign to that measured at 1.17 ev, which is sensitive to direct absorption effects as well. Thus, if the results of the measurement of free-carrier, intervalence-band, and indirect absorbance at 0.8 ev could be extrapolated to 1.17 ev, we would conclude that the change in absorbance due to these processes is smaller in magnitude and opposite in sign to that caused by the saturation of direct absorption coefficient. However, we would also conclude that the change in the combined free-carrier, intervalence-band, and indirect absorbance is of sufficient magnitude to substantially slow the saturation of the total absorbance at 1.06 μm with increasing carrier number. However, care must be taken when extrapolating absorbance measurements at 0.8 ev to 1.17 ev. Free-carrier and intervalence-band absorption coefficients are expected to decrease with increasing photon energy for a given (large) carrier density[17] while, according to Ref. 16, the Coulomb-assisted indirect absorption coefficient should increase.

The experiments shown schematically in the inset of Fig. 6 were repeated for a sample temperature of 295 K. Similar results were obtained. For the maximum carrier densities achieved at room temperature, the decrease in absorbance at 1.06 μm was 2.2 and the increase in absorbance at 1.55 μm was 1.8.

The results of excite-probe experiments that measure the temporal evolution of the change in absorbance at 1.55 μm are presented in Fig. 7. In this experiment, the sample was irradiated by an optical pulse at 1.06 μm containing roughly 2×10^{15} quanta and was probed at various delays by a weak pulse having a wavelength of 1.55 μm. The results of

the probe measurements at 1.55 μm are similar to those obtained by Auston et al.[4] However, these authors stated that they performed their measurements at excitation intensities such that the absorption of the excitation pulse was linear. Our experiments are clearly performed in the nonlinear region. In addition, the measurements of Auston et al.[4] were performed on a 300-μm-thick sample, our sample was 6-μm-thick.

The measurements presented in Fig. 7 indicate that free-carrier, intervalence-band, and indirect absorption can be significant at the carrier densities encountered during the excite and probe experiments at 1.06 μm presented here. Auston et al.[4] attribute this decrease of the probe pulse absorbance at 1.55 μm with delay to a decrease in free-carrier absorption caused by a temporal decay in carrier density due to Auger recombination. The present experiments only allow the measurement of the change in the combined free-carrier, intervalence-band, and indirect absorbance, and they do not provide for a convenient separation of their individual contributions. These measurements were performed for a sample temperature of 295 K, as well. The results are similar to those of Fig. 7. It is important to note that we observe a strong temperature dependence in the rise in probe transmission at 1.06 μm (see Fig. 4 or Fig. 5); however, we do not observe a similar strong temperature dependence at 1.55 μm. We believe this is a further indication that indirect, free-carrier, and intervalence-band processes do not dominate the rise in probe transmission at 1.06 μm.

VI. SUMMARY AND CONCLUSIONS

The measurements by Smirl et al.[5] of the transmission of a 1.06-μm-probe pulse as a function of time delay after an intense 1.06-μm-excitation pulse have been carefully repeated for sample temperatures of 100 K and 295 K. The rises in probe transmission for the two temperatures have been compared to a calculated integration curve, assuming an optical pulsewidth of 10 psec. We conclude from this comparison that the rise in probe transmission at 295 K is indistinguishable from the integration curve, but that the rise at 100 K is much slower than either the integration curve or the rise at 295 K and is not an artifact of the measurement technique.

The transmission of a thin germanium sample at 1.55 μm and 1.06 μm has been measured as a function of optically-created carrier densities. Over the range of densities encountered in these experiments, the absorption versus density relationship at 1.17 ev does not exhibit a minimum. Thus, a temporal decay of carrier density alone cannot be combined with this absorption versus density relationship to account for the rise in probe transmission at 1.06 μm exactly as suggested in Sec. III. In addition, these measurements indicate that the combined free-carrier, intervalence-band, and indirect absorbance changes are opposite in sign and smaller in magnitude than the changes caused by saturation of the direct absorption. As a result, we believe that the decrease in absorbance at 1.06 μm with increasing carrier number is dominated by a saturation of the direct absorption coefficient; however, the rate of this decrease in absorbance is slowed by the contributions of these "other" processes that are opposite in sign.

In addition, the absorbance of a 1.55-μm-probe pulse has been measured as a function of time delay after an intense 1.06-μm-excitation pulse. The 1.55-μm-probe absorbance decays by approximately 0.8 in the first 100 psec following excitation, corresponding to a transmission increase of a factor of approximately 2. This represents a significant decay in the combined free-carrier, intervalence-band, and indirect absorbance during this period. Contrary, however, to measurements of the probe rise at 1.06 μm, the decay of probe absorbance at 1.55 μm exhibited no strong dependence on sample temperature.

As a result of the present measurements, the authors feel that free-carrier, intervalence-band, and Coulomb-assisted transitions combined with Auger recombination are not the mechanisms dominating the rise in 1.06-μm-probe transmission at 100 K. The contributions of these processes are significant, however, and they must be accounted for by any successful model. Unfortunately, the present measurements yield no direct information concerning carrier distribution temperatures or energy relaxation rates, and the question of attributing the rise in 1.06-μm-probe transmission to a cooling of a hot carrier plasma created by the excitation pulse remains unresolved.

Finally, we emphasize that we are aware that our measurements at 1.55 μm (0.8 ev) monitor the free-carrier, free-hole, and indirect absorption at an energy different from that of the experiments we are attempting to interpret (rise in probe transmission at 1.17 ev); however, we believe these experiments give the best available indication of the possible importance of these processes at the high optically-created carrier densities encountered in excite and probe studies at 1.06 μm.

The authors wish to acknowledge their debt to Dave Auston and Sigrid McAfee of Bell Laboratories for suggesting and sharing the recombination model presented in this paper and for their helpful discussions.

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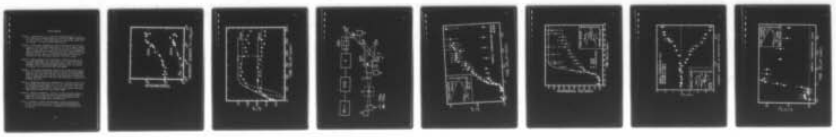

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FIGURE CAPTIONS

Figure 1: Transmission of a 5.2-μm-thick germanium sample at 1.06 μm as a function of incident quanta at 1.06 μm for sample temperatures of 100 K and 297 K. The dashed lines are theoretical curves from Elci et al.[7] The data are from Smirl et al.[5]

Figure 2: Probe pulse transmission versus delay between the excitation pulse at 1.06 μm and the probe pulse at 1.06 μm for sample temperatures of 100 K and 297 K. The data are plotted as the normalized ratio of probe pulse transmission to excitation pulse transmission T_P/T_E in arbitrary units. The dashed lines are theoretical curves from Elci et al.[7] The solid lines are theoretical curves from van Driel.[15] The experimental data are from Smirl et al.[5]

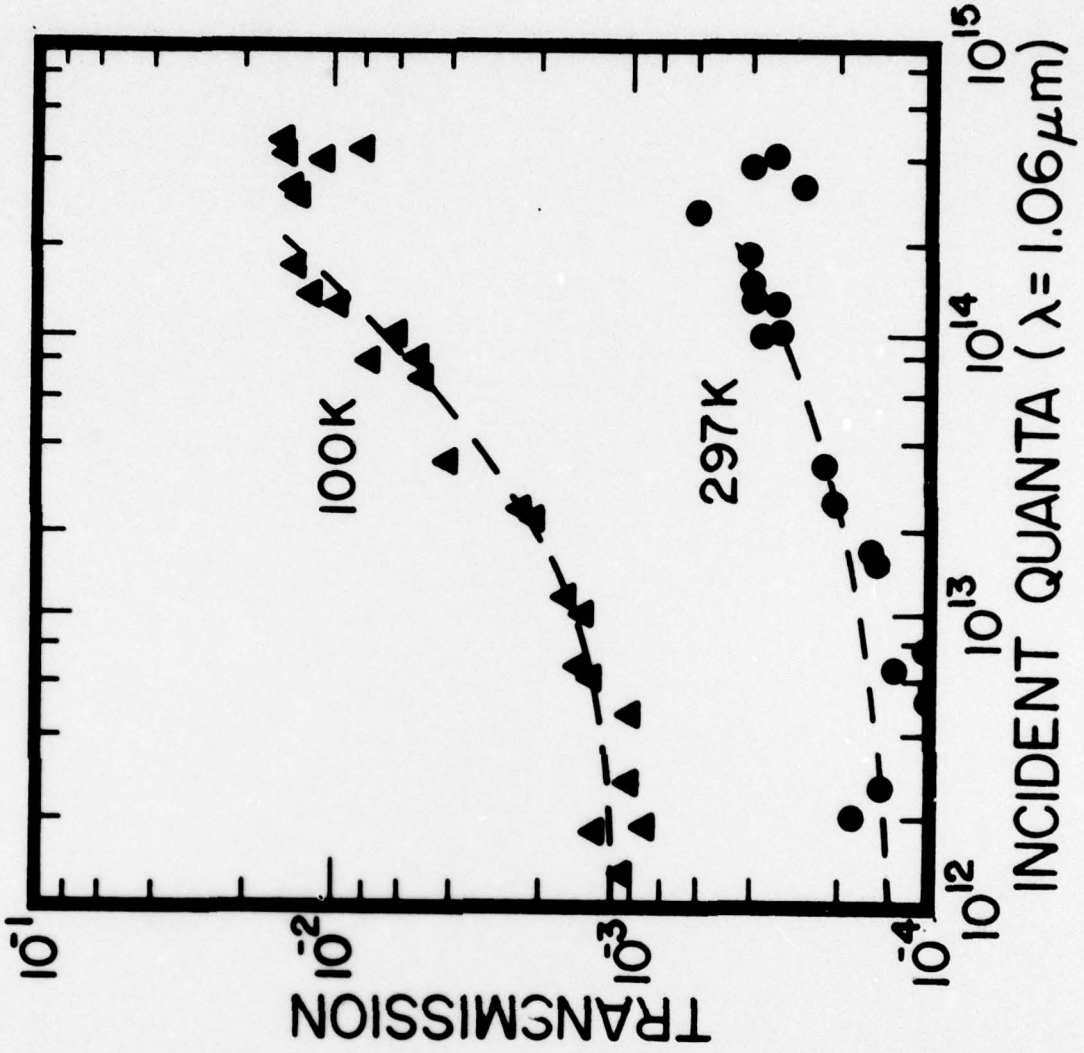
Figure 3: Block diagram of the experimental configuration for excite and probe measurements at 1.06 μm and 1.55 μm, where MLL denotes the mode-locked laser, EOS the electro-optic switch, A the laser amplifier, SRS the stimulated-Raman-scattering cell, M a mirror, D a detector, L1 and L2 lens, and S the sample.

Figure 4: Probe pulse transmission versus delay between the excitation pulse at 1.06 μm and the probe pulse at 1.06 μm for sample temperatures of 100 K and 295 K. The data are plotted as the normalized ratio of probe pulse transmission to excitation pulse transmission T_P/T_E in arbitrary units. The error bars represent one statistical standard deviation.

Figure 5: Normalized probe pulse transmission in arbitrary units versus delay between the excitation pulse at 1.06 μm and the probe pulse at 1.06 μm for sample temperatures of 100 K and 295 K. The solid line represents a theoretical integration curve assuming Gaussian-shaped optical pulses of 10 psec width (FWHM).

Figure 6: Change in absorbance, $-\ell n(T/T_0)$, of the germanium sample at 1.06 μm and 1.55 μm as a function of incident excitation pulse energy at 1.06 μm, where T_0 is the linear transmission of the sample at the wavelength under consideration.

Figure 7: Change in probe pulse absorbance, $-\ell n(T/T_0)$, versus delay between the excitation pulse at 1.06 μm and the probe pulse at 1.55 μm, where T_0 is the linear transmission of the probe pulse at 1.55 μm.


TRANSMISSION
10^{-1}
10^{-2}
10^{-3}
10^{-4}
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297K
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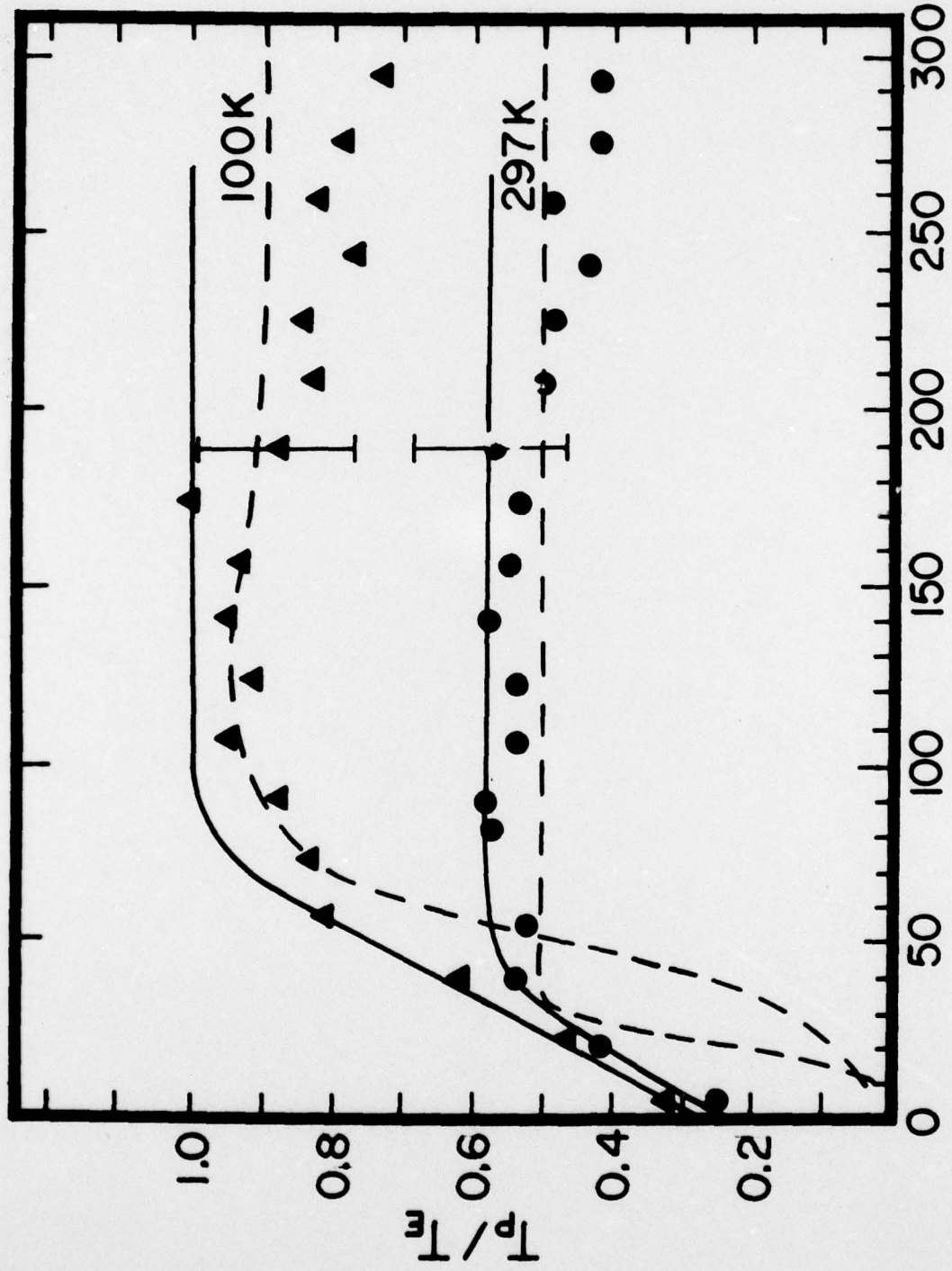
10^{13}
10^{14}
10^{15}
INCIDENT QUANTA (λ= 1.06μm)


100K
297K
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0.2
T_p/T_e
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50
100
150
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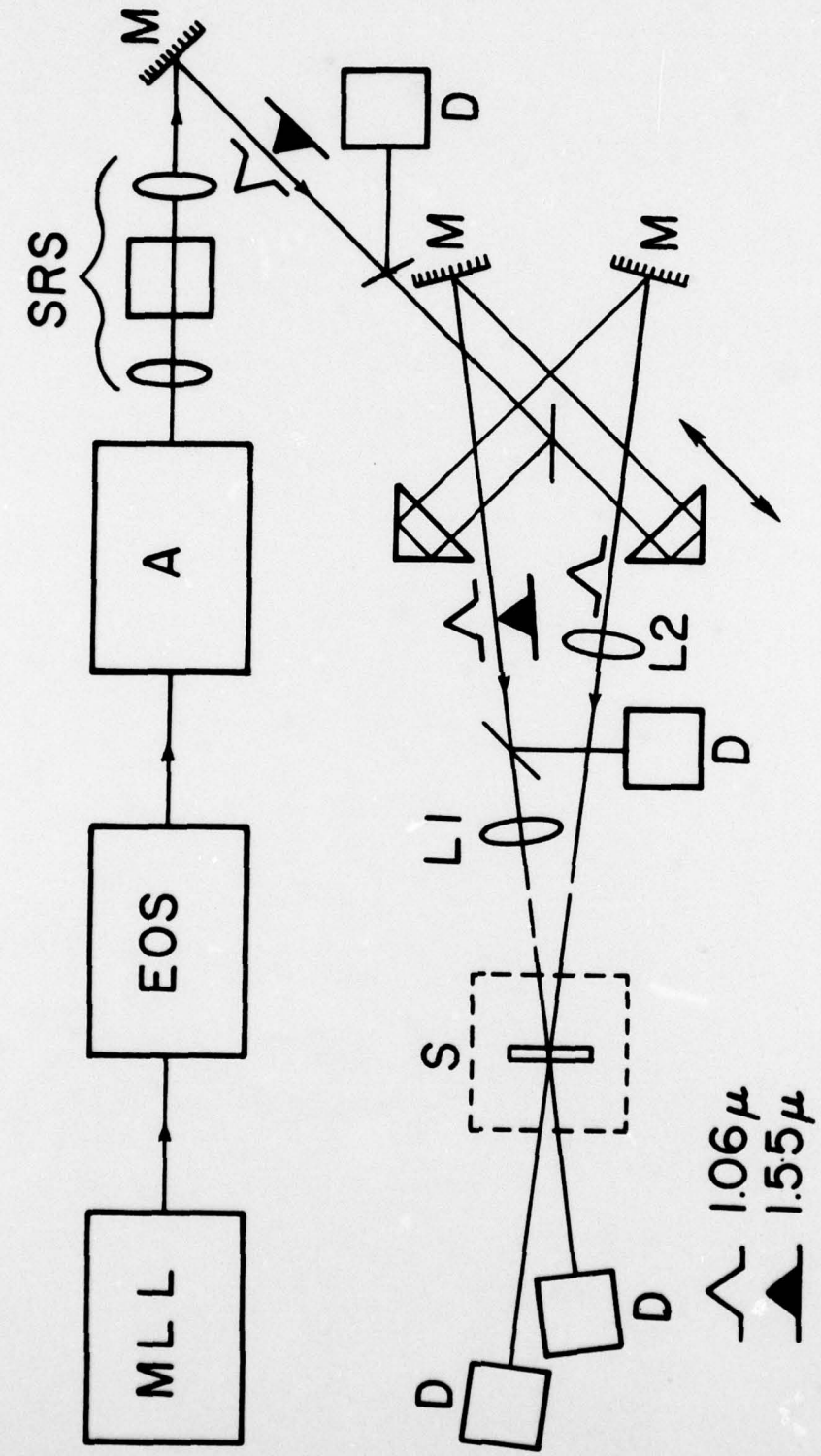
TIME DELAY (psec)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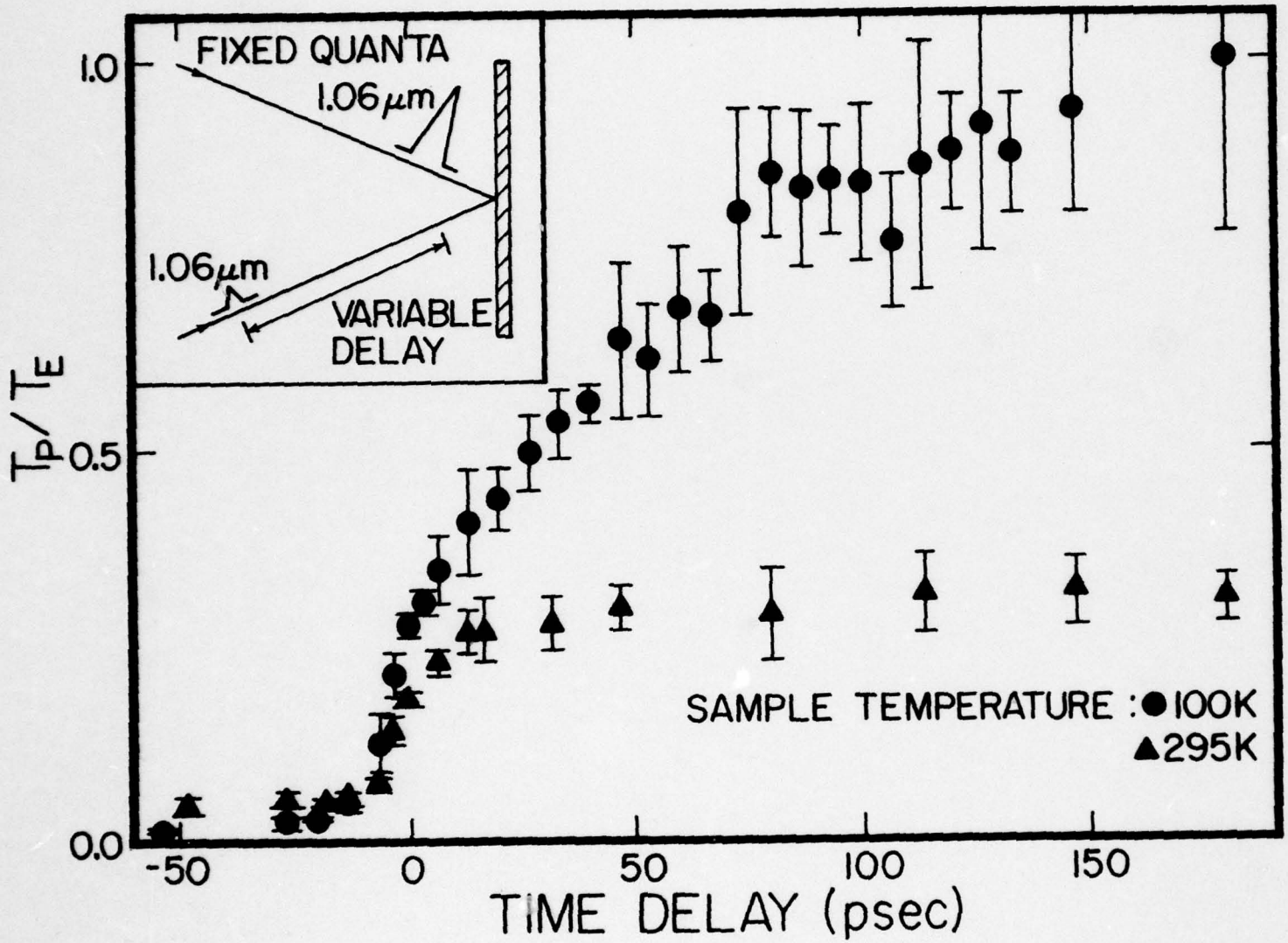
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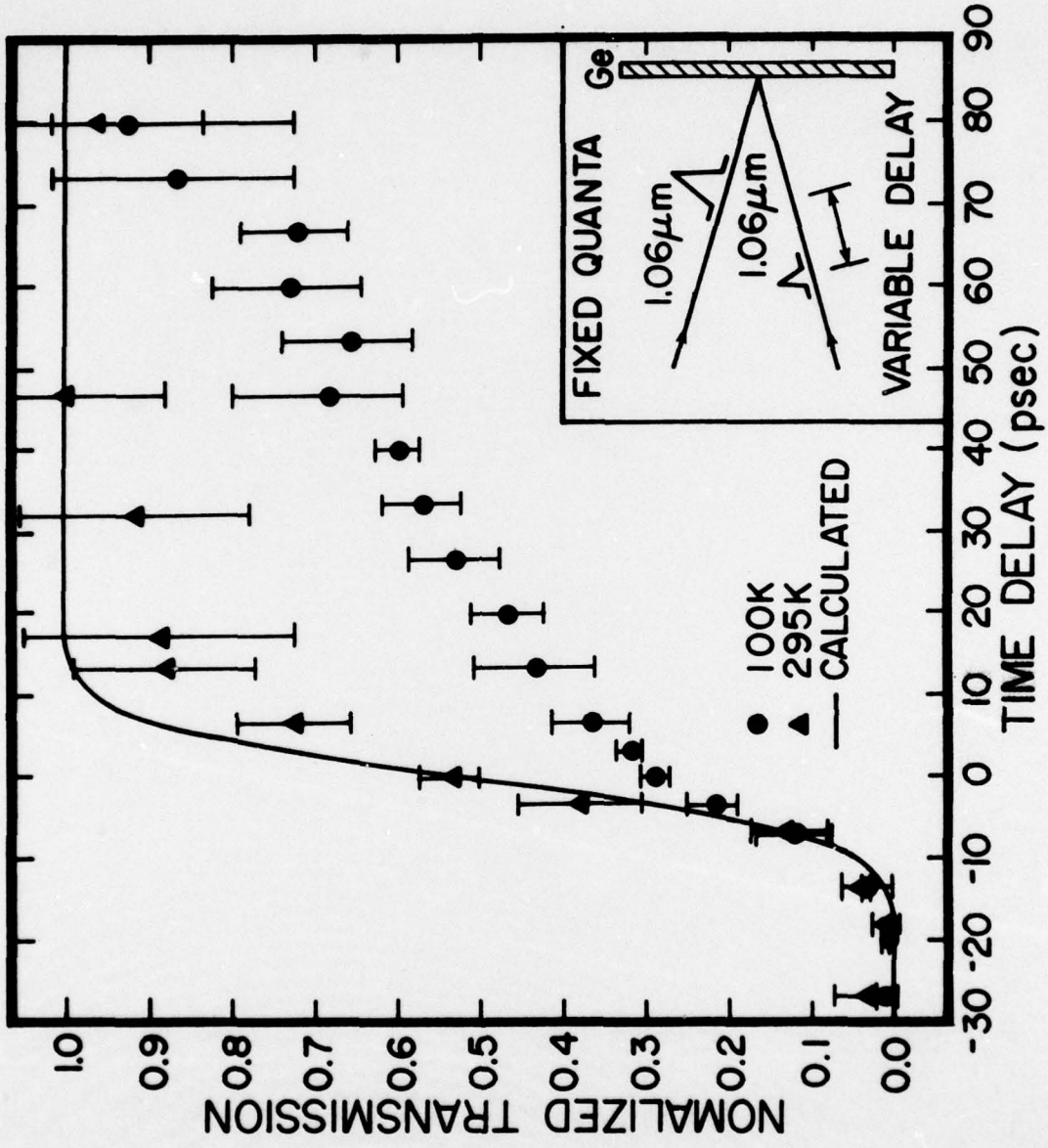
▲ 295K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0.5
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0.2
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-30 -20 -10 0 10 20 30 40 50 60 70 80 90
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Ge
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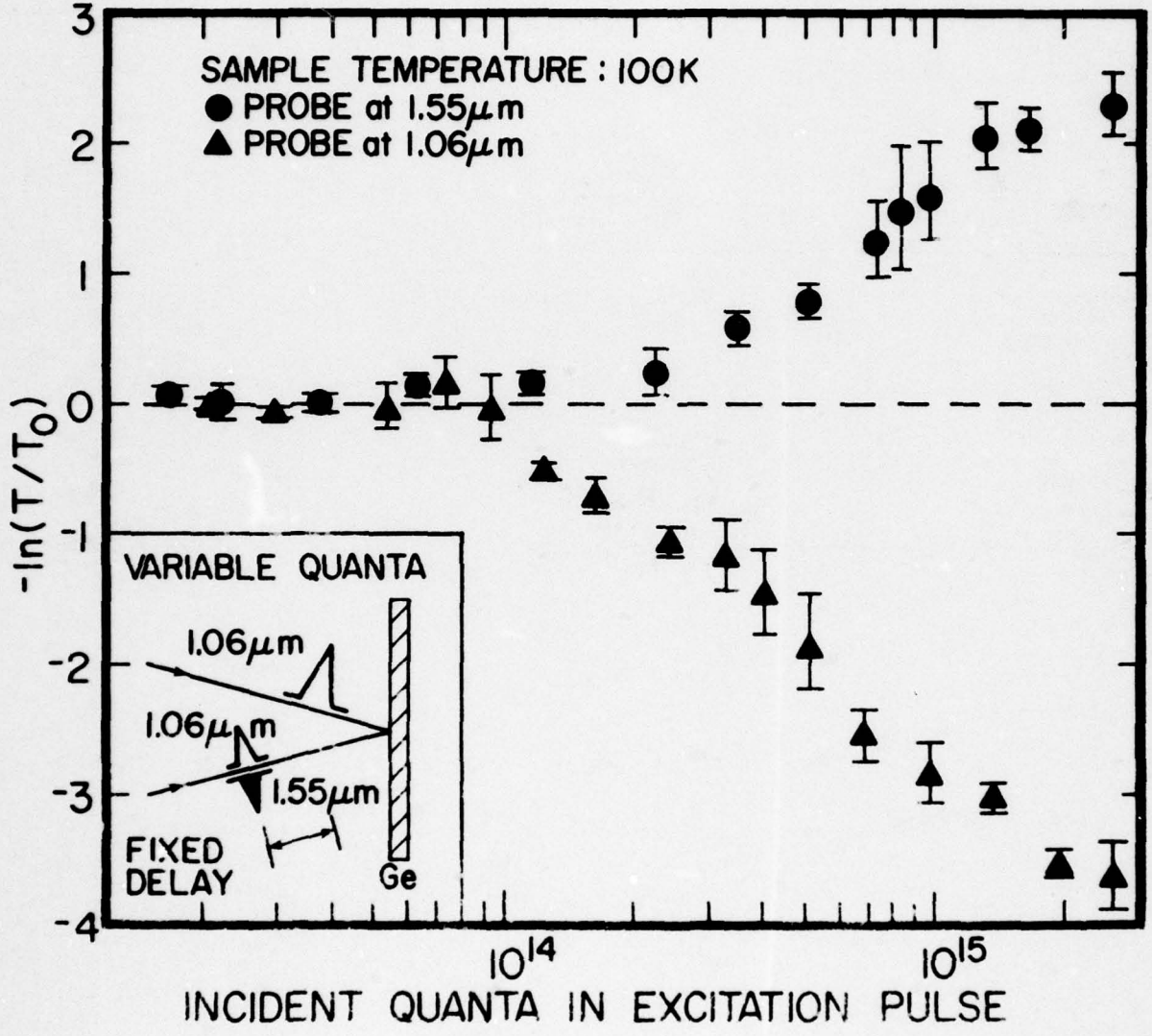
VARIABLE DELAY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-3
-4
VARIABLE QUANTA
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FIXED DELAY
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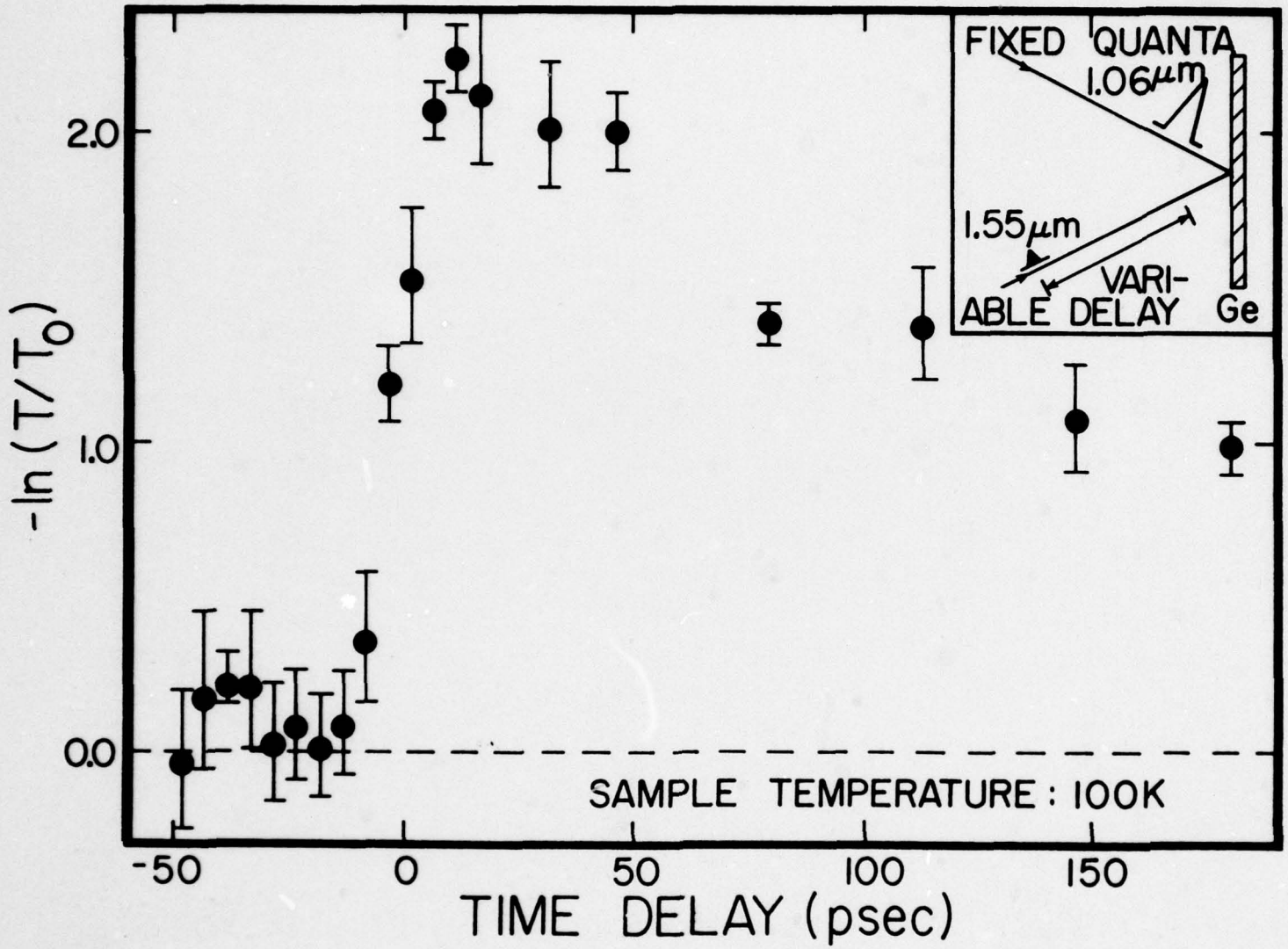
10^14
10^15
INCIDENT QUANTA IN EXCITATION PULSE


FIXED QUANTA
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Ge
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-ln (T/T0)
2.0
1.0
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-50
0
50
100
150
TIME DELAY (psec)